### WASTE CALCINER OFF-GAS SYSTEM

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### ABSTRACT

Provisions for decontamination of the spent fluidizing air and other calciner off-gases presented major problems in the design of the pilot plant now being constructed at the National Reactor Testing Station for converting ICPP aluminum nitrate wastes to solids. In passing through the fluidized bed, the fluidizing air entrains radioactive particulate matter, a volatile radio-active fission product, and the oxides of nitrogen formed in the calcination reactions. This paper describes the off-gas cleaning system adopted for the Waste Calciner and gives some of the process considerations that entered into its design. Expected performance is given for the various types of air cleaning equipment employed. Particular emphasis is placed on the novel operations involved in the use of silica gel as a medium for adsorbing ruthenium and trapping dust. A new fixed bed cleaning method is described for removing the dust and ruthenium trapped in the silica gel. The operability of this and other ruthenium adsorber operations is substantiated by the results of a research program designed to test the mechanical performance of this equipment.

### INTRODUCTION

Safe and economical long term storage of fission product bearing wastes is the goal of research now being conducted to develop processes for converting liquid wastes to highly inert solids. Existing aqueous processes for recovering fissionable materials from spent reactor fuels yield radioactive waste liquids which must be retained in expensive permanent storage. These liquid storage containers, and probably any type liquid storage which can be devised, are of questionable durability to withstand the earthquake, corrosion and other damage which might occur in the 300 to 400 year confinement period needed to reduce the radioactivity to levels generally acceptable for discharge to man's environment. The wastes are potentially more safely and economically stored in the solid rather than the liquid state due to the reduced mobility of the fission products, and the reduced volume of stored material. Argonne National Laboratory and Phillips Petroleum Company have performed the research work for a fluidized bed calcination process to convert to a solid the aluminum nitrate type waste liquid generated in the Idaho Chemical Processing plant. The Waste Calcination Facilities now being constructed at the ICPP will serve as the pilot plant for further development of this process and to obtain engineering data for future fluidized bed waste calcination plants.

Fluidized bed calcination is an excellent means of converting the ICPP aluminum nitrate type waste to a granular, pneumatically-transportable solid, but in this case, it is also the source of a severe gas cleaning problem. In this operation particles of the fission product bearing calcined product (Al<sub>2</sub>O<sub>3</sub>) are entrained in the fluidizing air and one of the radioactive fission products (ruthenium) is volatilized. Decontamination factors of 10<sup>4</sup> for the dust and 10<sup>3</sup> for the ruthenium are minimum requirements for discharge to the ICPP stack when calcining the 200 day out-of-reactor MTR wastes which serve as the design basis for the Waste Calcination Facilities. This gas cleaning problem is complicated by the presence of a high level of radioactivity. Simple dependable equipment is needed to minimize the frequency of the costly operations involved in the maintenance of such radiochemical plants and to facilitate the decontamination operations preparatory to direct maintenance.

### OFF-GAS SYSTEM

Two major processing requirements governed the design of the calciner Off-Gas System. One was the need to reduce the calciner off-gas dust concentration by a factor of  $10^4$  and the other to reduce the ruthenium concentration by  $10^3$ . Commercial equipment was available for most of the dust removal services. This was not so for ruthenium. The limited amount of published data on ruthenium succeeded only in confirming our belief that we were dealing with one of the most capricious elements known to man. Further, ruthenium is present only to the extent of 0.11 grains  $RuO_L/1000$  CF of calciner off-gas. The very effective caustic scrubbing method was not applicable to this problem as large volumes of caustic

would also react with the oxides of nitrogen present in the off-gas to give a neutralized waste having a volume larger than the acid waste originally fed to the plant. Water scrubbing would yield a mixture of ruthenium and nitric acid from which the ruthenium could be separated only by extreme and costly processing. These difficulties led to the search for a solid material to which ruthenium would adhere and preferentially separate from the water, oxides of nitrogen, and other components of the off-gas. This type separation was suggested by the well-known "plating-out" tendency of RuO4 on many types of surfaces. Phillips Petroleum investigated a number of these substances as well as commercial adsorbents. As a result of their laboratory research, silica gel was selected for use in the Waste Calcination Facilities.

The off-gas system which was developed to remove both dust and ruthenium from the calciner off-gas is shown in Figure 1. Dust which is collected in the first two devices - calciner cyclone and venturi scrubber - is returned to the calciner fluidized bed. Most of the particles collected in this equipment agglomerate and pass out of the calciner with the calcined product to solids storage.

Water circulating to the venturi scrubber and quench tower absorbs oxides of nitrogen and ruthenium until equilibrium concentrations of these two gases build up in the water. Thus, there is no net recovery of ruthenium or oxides of nitrogen in this part of the plant. Oxides of nitrogen pass on through the system and out to the ICPP stack. Ruthenium is retained in the ruthenium adsorbers.

### Process

Aluminum nitrate type ICPP waste enters the 4-foot diameter calciner and is sprayed into the hot fluidized bed through atomizing nozzles. The manner in which the atomizing nozzles are operated determines to a great extent the dust loading in the gas leaving the bed. Fluidizing air enters beneath the bed through a concave, perforated grid. The heat required to bring the feed to the  $400^{\circ}$ C calcination temperature and to support the reaction is furnished by heated

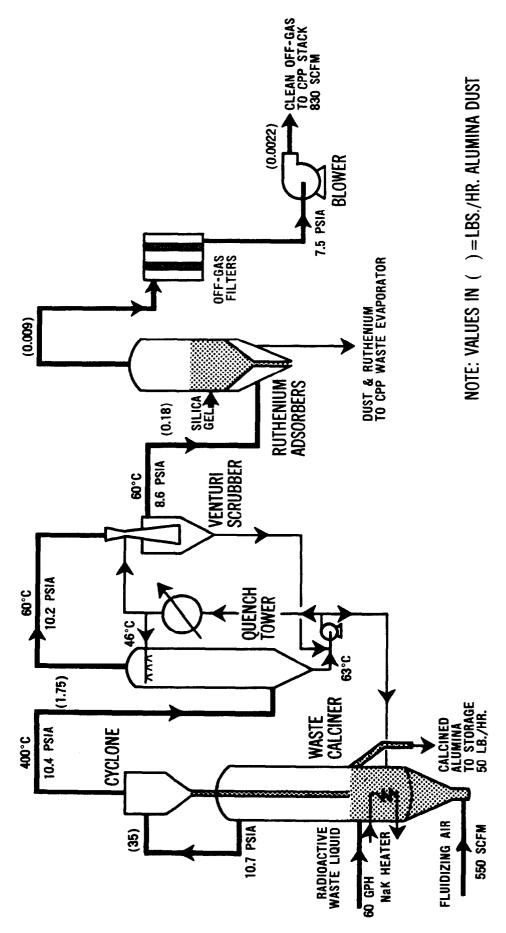


Fig. 1 --- Waste calciner off-gas system.

NaK circulating through an exchanger located in the fluidized bed. Fluidizing air is preheated in the fired heater which is used to heat the circulating NaK to  $760^{\circ}$ C. The composition and quantity of the calciner off-gas are shown in Table I.

The calciner cyclone is a highly efficient unit consisting of 16 parallel cyclones of 4-inch diameter. Fines collected in this unit flow by gravity to the fluidized bed through a dip leg.

The second dust removal device in the off-gas train is a venturi scrubber. This is preceded by the quench tower which cools the calciner off-gas to the venturi scrubber operating temperature. The large temperature spread between the calciner and the venturi makes the quench tower necessary - for a smaller temperature difference, sufficient cooling could be effected in the venturi alone. No packing is provided in the quench tower, an empty  $5\frac{1}{2}$ -foot diameter by 9-foot vessel with 4 spray nozzles for atomizing the quench water. A commercial venturi scrubber and disengaging vessel are used for the dust removal service. Water is pumped to the venturi throat at a rate of about 20 gal./1000 CFM of off-gas.

Equilibrium concentrations of nitric acid and ruthenium, and a controlled concentration of dust build up in the water circulating to the quench tower and

TABLE I

CALCINER OFF-GAS COMPOSITION

Component	Vol. %
02	16.4
N <sub>2</sub>	58.5
NO-NO <sub>2</sub>	3.5
H <sub>2</sub> O	21.6

CFM ( $\bullet$  400°C and 10.7 PSIA) = 2840

venturi scrubber. The equilibrium nitric acid concentration is 40 weight per cent. Total Al<sub>2</sub>O<sub>3</sub> content of the liquid is maintained at about 10 mg/ml by a purge of 20 GPH which is returned to the calciner. Water circulation to the quench tower is 3770 GPH and to the venturi 1680 GPH. Heat transferred to the quench water in cooling the off-gas is rejected to the plant cooling water system through a shell and tube exchanger.

Dust and ruthenium are trapped in the silica gel beds of the ruthenium adsorbers. These beds are periodically regenerated with water to remove the accumulated dust and adsorbed ruthenium.

Final cleaning of the off-gas is accomplished in three parallel units, each consisting of a Fiberglas prefilter followed by a glass-asbestos final filter. The prefilter has an NBS discoloration test efficiency of 30 - 35 per cent and the final filter a DOP efficiency of 99.97 per cent. The filter face velocities are 180 ft./min. during normal operation, 220 ft./min. during adsorber regeneration, and 270 ft./min. during filter replacement. Filter life is expected to be 600 to 1000 hours.

Filter replacement is carried out remotely due to the high radiation intensity of the accumulated dust. Two of the three parallel units are left on stream while the third is being replaced. A filter is shown in the on-stream position in Figure 2. The filter elements are contained in a disposable housing which is equipped with internal gas inlet and outlet valves (not shown in Figure 2). When a filter is to be removed these internal valves and the inlet and outlet valves in the piping are closed manually from the operating corridor by means of the valve operators and handles shown. The flanges are disconnected and the filter is lifted into the lead shielded (6-inch thick) transport cask. The cask is then lifted from the cell and removed to a burial ground where the filter container is deposited.

A 16 stage centrifugal blower provides the 6.9 PSIA pressure differential needed to pull the off-gas through the cleaning equipment and maintain all portions of the system under vacuum. It is driven by a 200 HP electric motor. The calciner and its off-gas system must be maintained at a negative pressure with respect to the cells so that any leakage will be fresh air into the processing equipment rather than contaminated air to the cell ventilation system.

Condensate formation in the off-gas leaving the venturi scrubber would cause damage in the silica gel (decrepitation), filters, and blower. A heater (not shown in Figure 1) raises the off-gas temperature to 66°C before it enters the silica gel. This temperature is maintained up to the filters by heating the cells housing the adsorber valve manifolds, adsorbers, and filter inlet piping manifolds.

### Performance of Off-Gas Equipment

Expected performance of the off-gas equipment is difficult to predict due to uncertainties as to the quantity and size of particles which will be generated in the calciner and a lack of data for operation of the elements of the off-gas system as an integrated unit. Efficiencies and particle size distribution were assumed for the design basis, using data obtained by Phillips in a 6-inch diameter calciner equipped with a cyclone and venturi scrubber and operated on a synthetic, non-radioactive feed. The design performance of the various elements of the off-gas system is shown in Table II.

### RUTHENIUM ADSORBERS

The results of the Phillips ruthenium decontamination studies suggested the use of a regenerable silica gel bed for removing ruthenium from the waste calciner off-gas. This work showed that silica gel would adsorb the ruthenium species generated in the laboratory under waste calcination conditions of temperature, pressure, and composition. When this synthetic calciner off-gas was cooled to around 60°C and passed through silica gel at a velocity of 1.0 ft./sec., or less, decontamination factors of  $10^3$  were obtained. The silica gel continued to remove ruthenium with this same efficiency after a total bed loading of 40 grams of ruthenium per cubic foot of silica gel. Fair desorption of the ruthenium from the bed could be obtained by washing with water.

Previous experimental work by the Bureau of Mines<sup>1,4</sup> and Sachsse<sup>3</sup> with moving coke bed filters, indicated that silica gel, when placed in the calciner off-gas system, would soon become clogged with dust. In fact, previous plant<sup>5</sup> and experimental experience with coke bed filters led to the consideration of using the ruthenium adsorbers as dust removal devices. In order to employ silica gel as a dust and ruthenium removal medium, it was first necessary to develop a simple, remote method for cleaning the bed of radioactive dust. The method of dust removal used in moving coke bed filters was considered much too complex

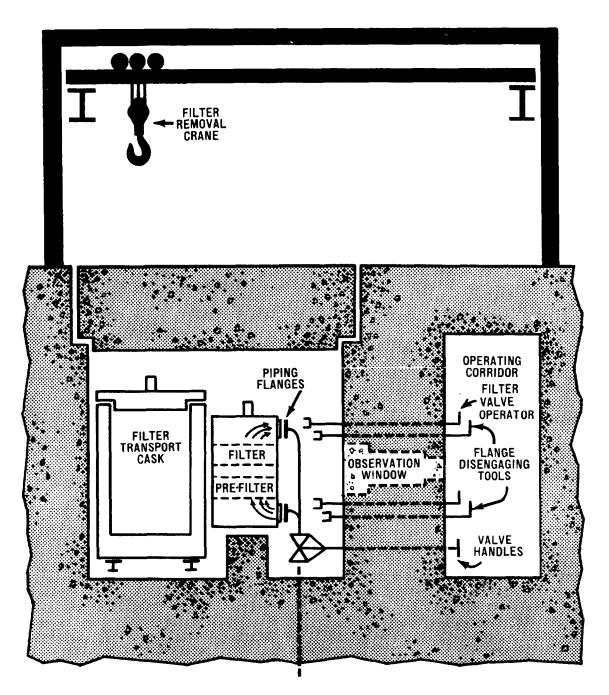


Fig. 2—Off-gas filter.

mechanically for use in a radiochemical plant. A ruthenium adsorber operating cycle was proposed and Fluor was asked to conduct a very specific research program<sup>2</sup> directed at determining the mechanical workability of the proposed scheme and developing alternative methods where needed.

TABLE II

DESIGN PERFORMANCE OF OFF-GAS EQUIPMENT

	Calciner Cyclone	Venturi Scrubber	Ruthenium Adsorbers	Off-Gas Filters
Inlet Dust Loading, Grains/1000 CF	1500	146	12	0.38
Design Dust Removal Efficiency, %	95	95	95	75
Pressure Drop, Inches H2O	8	40	6 to 14	1 to 4

### INLET PARTICLE SIZE DISTRIBUTION, WT. %

Size, Microns			
> 147	77-3		
20 - 147	6.0		
2.9 - 20	6.6	12.8	17 (>1.24) -
0.6 - 2.9	7.0	58.8	18 (0.6-1.24)
0.2 - 0.6	2.8	25.6	51 -
< 0.2	0.3	2.8	14 -

### Operation of Ruthenium Adsorbers

Out of the Phillips and Fluor research, a method was evolved for operating the ruthenium adsorbers. This is shown schematically in Figure 3.

### 1. Adsorbing and Filtering

Four 7-foot diameter adsorbers are being installed in the Waste Calcination Facilities. The filtering/adsorbing bed consists of 6 to 12 mesh silica gel supported on a conical grating which is sloped at 30° from the horizontal to permit emptying the silica gel by gravity flow to the 8-inch diameter fluidizing leg. The bed varies in depth from 36 to 62 inches. Restrictive orifice distributors are placed below the bed to compensate for this difference in bed height.

Figure 3A shows one of the adsorbers in the on-stream position. Three of the four adsorbers are on stream at any one time, while the fourth is being regenerated, or held in standby. With three adsorbers on stream, the off-

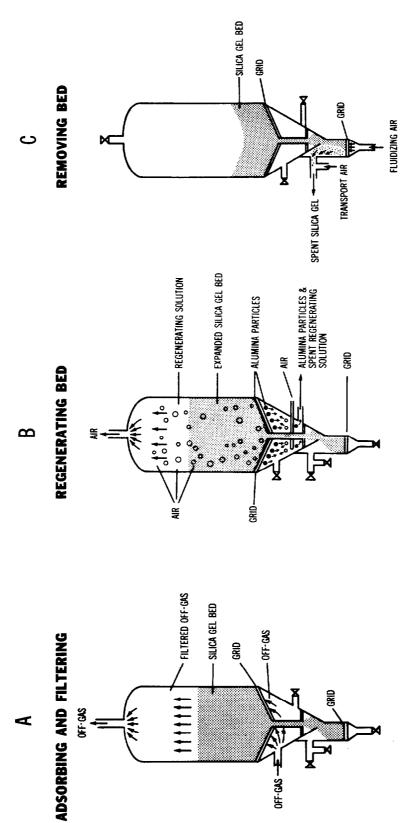


Fig. 3 -- Operation of ruthenium adsorbers.

gas superficial velocity through the bed is 0.33 feet per second. At this velocity, the pressure drop through the clean bed is 4-inches of water and through the distributing orifices 1.7-inches.

The laboratory work shows that ruthenium and dust both collect almost quantitatively in the lower 25 to 30 per cent of the bed. During plant operations, the build-up of dust on the bed rather than the degree of ruthenium adsorption will probably determine the length of time between regenerations, estimated to be 10 to 30 days.

### 2. Regenerating

The regenerating operation is shown in Figure 3B. Ruthenium removal is effected by washing the bed with three volumes of water. During the second wash, the water level is held above the silica gel bed and air is admitted below the grid through a sparger at a superficial velocity of 0.15 ft./sec. The air expands the silica gel, which has a specific gravity slightly above that of water, and causes a mild agitation of the granular particles. The smaller and heavier alumina particles become disengaged from the silica gel during this operation and fall to the bottom of the adsorber where they are swept out by the circulating water and are eventually pumped to the ICPP process waste evaporator. There is some indication that a small portion of the alumina particles adhere to the silica gel in a manner which is not detrimental to subsequent off-gas flow through the bed (i.e., pressure drop does not increase).

### 3. Drying

After regeneration, the silica gel bed is dried to 25 weight per cent contained water - the equilibrium water content of the bed when in contact with the calciner off-gas. Because of the possibility of contamination, the air used in drying is passed through the adsorbers which are on stream and into the off-gas filters.

### 4. Removing Bed

Should a silica gel bed become spent with respect to ruthenium removal or if it becomes necessary to perform maintenance in or on a contaminated adsorber, provisions are made for transporting the radioactive silica gel to the calciner product storage. As shown in Figure 3C, spent silica gel is pneumatically transported to the solids storage containers by a fluidizing leg at the bottom of the adsorber. The relative rates of the fluidizing and transporting air streams is determined by the pressure drop observed in the transport line.

### Experiments to Evaluate Mechanical Performance

Evaluation of the mechanical performance of the ruthenium adsorbers was conducted in two parts. The first, bench scale tests, was undertaken to develop a simple method of removing accumulated dust from the silica gel. The second, one-third scale tests of the proposed adsorber, was undertaken to determine the effects on the silica gel of repeated saturation, washing, and drying operations, and to check the proposed schemes for replacing contaminated silica gel.

### 1. Bench Scale Tests

The bench scale tests were conducted in a 6-inch diameter metal cylinder containing a 23-inch bed of silica gel. The plant filtering operation was simulated in this apparatus by subjecting the bed to air containing fine

alumina dust and water vapor in about the same concentrations as are expected for the plant ruthenium adsorber feed. The alumina dust, a fine cut from a laboratory cyclone, was made by grinding some of the product of the Phillips 6-inch calciner. A rough approximation of the particle size distribution of this dust was obtained by counting the various size particles observed under a microscope:

Particle Size Microns	Observed Particle Distribution
> 10	10
5 - 10	30
1 - 5	50
<b>4</b> 1	10

The effectiveness of the new dust removal technique is illustrated by Table III which shows that only a short period of mild air agitation is needed to return the bed to its clean pressure drop. In this experiment emphasis was placed on using small quantities of air for agitation as all effluent regeneration air from the plant adsorbers is considered contaminated, and thus it must be handled by the off-gas filters and blower.

This procedure - whereby the bed is flooded with water and air is bubbled through the bed and the water - does not appear to result in carrying dust into the bed. Visual inspection of the bed did not reveal any significant dust penetration due to the cleaning procedure.

Although the bench scale test was intended primarily for developing a method of cleaning the silica gel, pressure drop buildup and efficiency data taken during this experiment give some idea of what might be expected when the plant goes into operation. These are shown in Table III and Figure 4. However, since the plant dust will probably contain a higher per cent of 1 micron and smaller particles, the ruthenium adsorbers are expected to be less efficient than the bench scale filters and the rate of pressure buildup will probably be greater than shown in Figure 4.

### 2. One-Third Scale Tests

These tests were conducted in a one-third scale model of the proposed ruthenium adsorbers. The plant regeneration operations were simulated in this apparatus by subjecting the silica gel bed to moist air (same concentration H<sub>2</sub>O as plant off-gas, first, then slowly increasing to bring silica gel to its maximum water holding capacity), to liquid water, and to dry air. Each complete cycle required about 56 hours. A long saturation period (about 40 hours) was found necessary to insure against the tendency of silica gel to decrepitate when contacted with liquid water. We found that silica gel containing even as much as 39 weight per cent water (40% is saturation point) decrepitated when contacted with liquid water.

After 30 cycles of 56 hours each we found the silica gel to be free flowing and readily removable from the test vessel by the fluidizing leg. A small increase in silica gel fines was noted but the quantity generated was insignificant in its effect on pneumatic transport. The bed pressure drop increased from 1.5 inches of water for the first cycle, to 2.0 inches for the 30th cycle. This is a tolerable increase in view of the 8 to 25 regenerations per year expected for each plant adsorber. No attempt was made

TABLE III

## TYPICAL RESULTS OF BENCH SCALE TESTS

### SILICA GEL DUST FILTER

	Run No.	2]	긔	계	ជ	77	77	<u>16</u>
	Length of run in equivalent plant operating days	13.5	13.5 19.5	22.5	23.5	7	26.5	56
	Pressure Drop, Inches H20							
	Start of run	2.0	1.5	1.5	1.4	1.4	1.5	1.5
e E	End of run	0.6	6.9	7.0	6.7	7.3	6.9	6.5
	After regeneration	1.5	1.5	1.4	1.5	1.4	1.5	1.6
	Regeneration							
	Duration of Air Flow, Min.	180	8	45	83	Ħ	9	9
	Air Superficial Velocity, fps	0.15	0.15 to 0.20-				[3 ↑	→ 0.15 to 0.20
	Regen. Solution Volume, % Bed Vol.	98						<b>→</b> 300
		•						

99.5 99.8 99.5

99.5

99.2 99.0 99.2

Filtration Efficiency, %

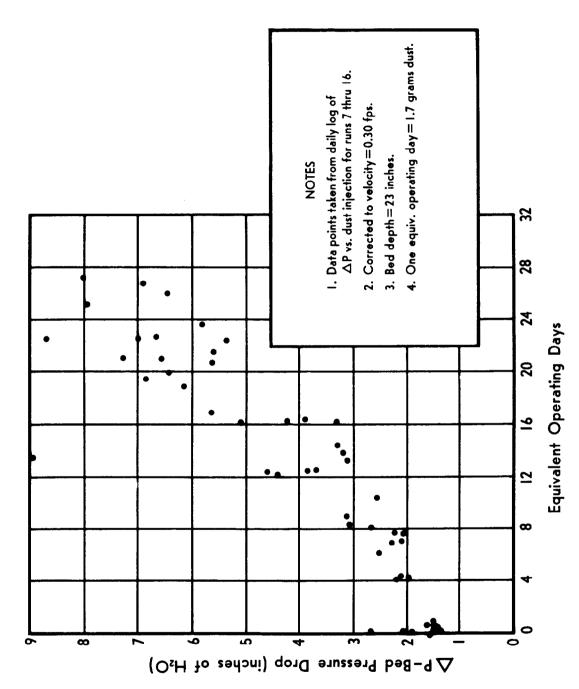


Fig. 4 -- Effect of dust accumulation on pressure drop through silica gel bed.

in the one-third scale test to employ the dust removal technique developed in the bench scale test.

The decrepitation characteristics and water holding capacity of the silica gel changed markedly after repeated regeneration cycles. After 10 cycles, silica gel dried with air at room temperature and humidity did not decrepitate when subjected to liquid water. After 30 cycles the silica gel could not be made to decrepitate even after drying to 245°C. The water holding capacity changed from 40 weight per cent adsorbed new to 33 per cent at 10 cycles, 30 per cent at 20 cycles, and 27 per cent at 30 cycles.

Pneumatic conveying was shown to be an impractical method of filling the adsorbers due to attrition of the silica gel. The plant adsorber filling operation was changed to gravity flow as a result of this test.

### CONCLUSION

The waste calcination pilot plant facilities described here should provide valuable information for improving the decontamination of radioactive aerosols encountered in many other operations as well as in the fluidized bed calcination of ICPP aluminum nitrate type waste. The research work performed by Argonne, Phillips, and Fluor provides a good basis for design of the pilot plant off-gas system. However, the data secured in operating the pilot plant will no doubt provide a firmer basis for the design of full scale plants and for improving the process.

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### THE ADSORPTION OF VOLATILE RUTHENIUM ON SILICA GEL

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### ABSTRACT

The removal of volatile ruthenium by solid adsorbers from an airnitric acid-water vapor phase was investigated. Silica gel was selected because it exhibited high affinity for ruthenium and the adsorbed ruthenium could be removed from the silica gel by washing with water. Decontamination factors obtained were in the range 100 to 1000 and total loadings as great as 58 grams of ruthenium per cubic foot of silica gel were experienced without obtaining a breakthrough of ruthenium.

### INTRODUCTION

Since the beginning of the atomic energy industry, it has been recognized that under certain conditions radioactive ruthenium can become an environmental hazard due to the formation of volatile compounds, which are readily dispersed in the environs. A laboratory study of methods for removing volatile ruthenium from calciner off-gas was made at the Idaho Chemical Processing Plant (ICPP) concurrently with a study of the fluidized bed calcination of radioactive wastes.

The problem arose from laboratory data which indicated that at  $400^{\circ}$ C, the operation temperature proposed for the fluid bed calcination of radioactive aluminum nitrate wastes, the ruthenium in the waste was almost totally volatilized, presumably as ruthenium tetraoxide. This volatile ruthenium contains appreciable amounts of ruthenium-106, and in young wastes, ruthenium-103. Inasmuch as the off-gas from the calciner is to be released to the environment, it was necessary to provide some means of removing ruthenium from the off-gas stream.

The information reported here is a review of the study made at the ICPF by Hanson, Newby, and Rohde(2) and a summary of the most recent results obtained by Anderson and Rhodes(1).

### Equipment

A simulated calciner off-gas was generated by continuously injecting a nitric acid solution containing ruthenium into a calciner maintained at 400°C as shown in Figure 1. In later models the U-tube adsorber was

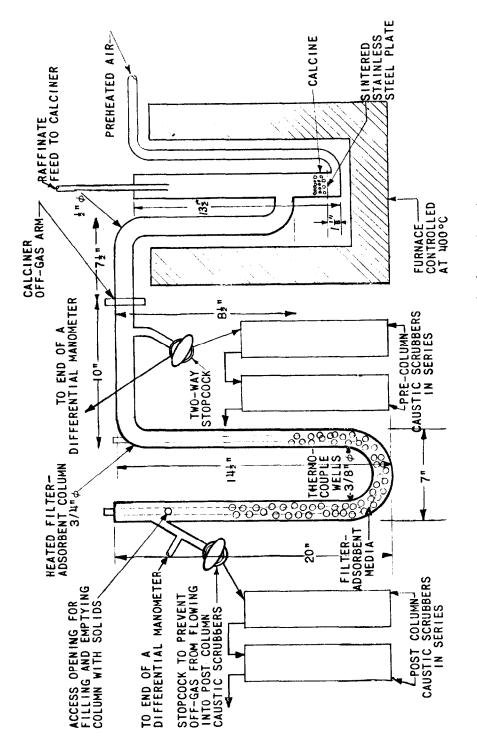


Fig. 1 -- Ruthenium calciner and adsorber equipment.

replaced by a straight tube. The feed solution contained only nitric acid and ruthenium traced with ruthenium-106. Aluminum nitrate was not included in the feed solution to avoid the formation of large amounts of solid with the subsequent problem of frequent cleanout of the calciner.

### Selection of Adsorbent

The method of approach was simply to generate the volatile ruthenium species and attempt to remove it from the off-gas stream by adsorption on a variety of solid adsorbents. The results of several short term laboratory tests with solid adsorbents are shown in Table 1. From these tests, which show ruthenium decontamination across the adsorber column, it is evident that several materials have a reasonably high affinity for the volatile ruthenium. Some of the adsorbents (alumina and molecular sieves) were eliminated because of physical degradation by the components of the off-gas and others were rejected (marlex, stainless steel, alumina) because the ruthenium was held so tightly by the adsorbent that it was impossible to remove the adsorbed ruthenium by washing with water or acid.

TABLE 1

THE ADSORPTION OF RUTHENIUM ON SOLID ADSORBENTS

130 cc of adsorbent at approximately 100°C

Adsorbent	Mesh Size of <u>Adsorbent</u>	Ruthenium <u>Decontamination Factor</u>
None (blank) Silica gel Activated Alumina	6-16 8-14	1.3 1200 2600
(commercial) Marlex	1/4" × 1/4" × 1/16"	325
Alumina (calcined at 400°C)	28 <b>-</b> 65	100
Linde Molecular Sieve Type 4A	1/16" pellets	46
Kaolin Cracking Catalyst	3/16" pellets	50
Porous Glass No. 7930	12-25	2500
Carpente <del>r 2</del> 0 Yorkmesh		178

### SILICA GEL STUDIES

Silica gel was chosen for further study because of its relatively high affinity for ruthenium, physical stability of the material under calciner operating conditions and ease with which the adsorbed ruthenium could be removed from the silica gel by washing. Figure 2 shows the elution of ruthenium from silica gel with various wash solutions. Water functioned equally as well as acids in removing the adsorbed ruthenium.

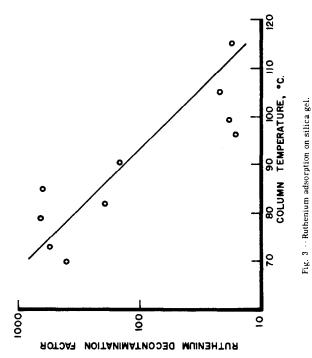
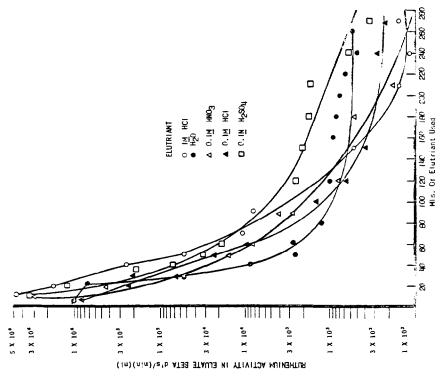


Fig. 2.—Elution of ruthenium from silica gel (27 cc of silica gel at 60 C).



### Effect of Vapor Velocity

Laboratory studies indicated that the superficial vapor velocity within the range 0.6 to 1.2 ft/sec had no noticeable effect on the adsorption of ruthenium; however, the lower vapor velocities very likely produced a steeper concentration gradient within the column.

### Effect of Column Length

Columns of 25 inches were of sufficient length to remove essentially all of the ruthenium under favorable operating conditions. When the length was reduced to 13 inches, the ruthenium DF was reduced to one tenth to one hundreth of the DF obtained with the longer column.

### Effect of Temperature

The effect of temperature was found to be very critical in its effect on ruthenium DF. Some of the data from one run are shown in Figure 3. In general, it appeared that operation of the adsorbing column just above the dew point was the most favorable temperature for adsorbing ruthenium.

### Determination of Loading Capacities

The determination of loading capacities during several adsorption-desorption cycles was attempted using a laboratory apparatus similar to that shown in Figure 1 with only minor modifications to permit continuous operation for several hundred hours. The results are shown in Table 2. A definite ruthenium breakthrough was not obtained in any of the runs. Run no. 1 was terminated due to inadequate temperature

TABLE 2

### RUTHENIUM ADSORPTION CAPACITY TESTS

110 cc of silica gel approximately 100°C Vapor Velocity 0.6 ft/sec

Run No.		Average Decontamination Factor	Loading Capacity grams Ru/cu.ft. of silica gel	Ru Eluted with Water at 60°C % of Total Adsorbed
1	140	23	22	55
2	233	326	40	75
3	209	394	24	.11
4	621	1013	58	48

control of the silica gel column. This poor temperature control resulted in a relatively low average ruthenium DF. Run no. 2 was terminated due to lack of sufficient feed solution. During run no. 3, the fluidizing air supply was interrupted and volatile ruthenium was generated for about 24 hours in the absence of fluidizing air with poor temperature control on the column. This malfunction probably accounts for the low recovery of ruthenium during the wash cycle. Run no. 4 was terminated because it appeared that the loading capacity was so high that dust loading in the demonstration fluid bed calciner would necessitate

# DIRECTION OF VAPOR FLOW

Fig. 4---Silica gel from ruthenium adsorber column.

washing the column long before the capacity of the silica gel for adsorbing ruthenium was exceeded.

Elution of the adsorbed ruthenium was moderately successful although an appreciable fraction of the ruthenium was retained by the silica gel as shown in Table 2.

The residual ruthenium in the column had no apparent detrimental effect on the adsorption of additional ruthenium. In fact, there appears to be a general trend of increasing ruthenium DF with continued use of the silica gel. Examination of the silica gel particles following the last run indicated a steep concentration gradient, Figure 4, with more than 98 per cent of the ruthenium being adsorbed in the first two-thirds of the column. Examination of the individual particles, which were blackened by the ruthenium, indicated that the ruthenium penetrated deeply into the particle, Figure 5. After sectioning an individual crystal, it was found that approximately 80 per cent of the total volume of the individual crystal was blackened by ruthenium.





Fig. 5—Silica gel from ruthenium adsorber column (Left, fragments of fractured particle; right, whole particle).

### SUMMARY

Based on the laboratory data, it appears that silica gel can be used to remove ruthenium from a nitrogen oxide-air-water vapor off-gas stream sufficiently well under controlled conditions to warrant its use as a means for decontaminating the off-gas from a fluid bed calciner. Ruthenium adsorber units are included in the demonstration calciner(3), which is presently under construction at the ICPP, and a hot run using full activity level off-gas will be made as soon as possible.

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### MEASUREMENTS OF ATMOSPHERIC DIFFUSION FROM AN ELEVATED SOURCE

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### ABSTRACT

Diffusion from an elevated source under lapse conditions was measured utilizing uranine dye as a tracer. Comprehensive measurements of temperature and vertical and horizontal wind fluctuations were made at several levels on a 150-foot tower. The meteorological parameters are correlated with the ground-level air concentrations in an effort to establish predictors directly from Eulerian meteorological data. The effects of wind direction shear in the vertical on the maximum ground concentration is discussed. Techniques of dispersal, collection, analysis and the results of particle size counting are included. Diffusion parameters measured from the dye concentrations are included and compared with those presently used at the National Reactor Testing Station.

### Introduction

In recent years atmospheric diffusion from ground-level point sources has been studied extensively at O'Neil, Nebraska through Project Prairie Grass in 1956 and the Great Plains Turbulence Project in 1953, sponsored by the Geophysics Research Directorate of the Air Force Cambridge Research Center. The results of these comprehensive ground measurements have been extended to elevated sources by Cramer / 1 /. Other investigators, Moses / 2 /, Stewart et al / 3 /, Singer / 4 /, and Smith et al / 5 / to mention a few, have studied diffusion from an elevated source, but their data have frequently been limited by the number of samples or they were concerned with a different aspect of the problem than the present discussion. It is believed that these experiments are the most comprehensive measurements of short range diffusion from an elevated source to date.

A total of 17 tests were performed from January to May 1959. The first test was discarded because of a slow wind shift during the period of the test resulting in three distinct concentration maxima. Uranine dye was used as a tracer for all tests as described by Robinson et al / 6 /. The characteristics of the dye are adequately described in the paper cited.

### Instruments and Measuring Procedures

One hundred high volume Staplex air samplers were used at various arcs at the NRTS Grid No. 3. Initially arcs were set up at distances of 300, 400, 600, 1000, 1800, and 3400 meters from the release point. After the eighth test the arc at 3400 meters was discontinued and a new arc at 150 meters was added. The 300, 400 and 600 meter arcs were widened from 40 to 60 degrees at this time. After Test No. 8, arc spacing of samplers was 60 on the 150, 300 and 400 meter arcs, 2.7 on the 600 meter arc, 1.7 on the 1000 meter arc and 1.80 on the 1800 meter arc. The odd spacing arose from the fact that the grid was originally set up for a source 100 meters from the first arc rather than 300 meters where the meteorological tower was located.

Hurlburt X-934-AH glass fiber filter papers supported by MSA 2133 all dust filters were used to collect the samples of uranine dye. The glass filter papers were too fragile to be inserted in the Staplex high volume air samplers without some supporting medium. The air flow through the filter pairs was approximately 15 cfm, a rate rather low for the high-volume type sampler. According to Smith and Suprenant [ 7 ], the Hurlburt glass paper combines the best features of particle penetration, flow rate, pressure drop and dust loading for our purposes. Our own investigation revealed background characteristics superior to all filter media tested except Millipore AA filters. Other filters tested included SS 589, MSA 2133, and Whatman No. 42. The background blank of the filters (i.e. clean filter washed in distilled water) was five times that of pure distilled water in the first batch of filters used (Tests Nos. 1 to 6); after that the succeeding batches of filters used exhibited the same background blanks as distilled water. We attribute this reduction to some change in the riber filler used in the Hurlburt paper. With the low background filters the lower detection limit, for 30-minute tests, of uranine dye was an air concentration of 1 x  $10^{-9}$  g/m<sup>3</sup>.

The dye solution was released at an elevation of 150 feet from the meteorological tower with an air flow of 500 cfm at 100 psi pressure. The dye solution was mixed at 3-1/2 pounds of dye per three gallons of water and was released over a 30 minute period. This resulted in a release rate of 0.88 g/sec of uranine dye. The writer is indebted to Dr. C. E. Lapple of the Stanford Research Institute for the dwsign of the adjustable nozzle used to disseminate the dye solution. The droplet size was not determined, but it was assumed to be small. The dye solution was red while the plume was brown, the natural color of dried uranine dye stuff. Lack of fallout was evidenced by particle size studies included later and no appearance of dye on the snow-cover close to the tower. During the time when the correct solution of dye and nozzle adjustment were being determined fallout did occur and was readily visible on the snow-cover surrounding the tower.

Samples were collected for particle sizing during two tests. The first set were collected on the glass fiber papers but were unusable because the smaller particles were imbedded in the filter and not visible in sufficient number to give a good distribution. This phenomenon was also mentioned by Smith and Suprenant [7]. The second sample was collected on Millipore AA filter paper and examined under a microscope using the oil immersion technique. The results are presented in Table 1.

A third sample was attempted but the plume divided and missed the electrostatic precipitator employed for this test and insufficient particles were collected for sizing. The results listed in Table 1 are regarded as preliminary until more evidence is obtained. There is a tendency toward decreased particle size at greater distance but no significance is attached at this time pending more sampling of particle sizes.

The exposed filters were collected in covered plastic petri dishes and washed in 20 ml of distilled water. Four ml of one molar phosphate buffering solution were added to bring the solution to a constant Ph value. The concentration was read by measuring the fluorescence with a Photovolt Model 540 Fluorescence meter. Investigation indicated that the fluorescence was sensitive to Ph but not to temperature so the samples were analyzed at room temperature. One other departure from the method outlined by Robinson et al / 6 / was made in that a square quartz curvette was employed. There was evidence that scratches on round glass test tubes caused a change in the reading of the meter for the same solution.

TABLE 1
PARTICLE SIZE DISTRIBUTION

Arc Distance (meters)	Median Size (microns)	Standard Deviation (microns)
600	1.0	1.80
600	1.42	1.87
600	1.35	2.67
1000	1.20	1.50

Meteorological instrumentation included Beckman and Whitley anemometers at 20, 75 and 140 feet on the tower, bivanes at 20 and 140 feet, and temperature elements at 5, 75 and 150 feet. An Aerovane was placed at 3 meters on the 1800 meter arc.

### Concentration Data

Figures 1, 2 and 3 illustrate the ground concentration data for three of the tests conducted. They are presented to illustrate the type of ground concentrations that occur with a release from an elevated source under unstable conditions.

Table 2 lists some of the pertinent data for the dye tests that are of particular interest to those using Sutton's  $\begin{bmatrix} 8 \end{bmatrix}$  diffusion equations. An explanation of the method of determining the parameters seems in order. The stability parameter n was measured from wind speed profiles where available but rounded off to either 0.20 or 0.25, whichever was closer. The actual range of n values was 0.17 to 0.26. The values for distance to maximum concentration ( $d_{max}$ ) and maximum concentration ( $\chi_{max}$ ) were obtained from a plot of the peak concentrations for each arc on log-log paper. The computed  $\chi_{max}$  was obtained from

$$\chi_{max} = \frac{2 q}{e \pi u h^2}$$

(1)

The vertical diffusion coefficient  ${\rm C_z}^2$  was determined from the equation for  ${\rm d_{max}}$ , i.e.

$$d_{\text{max}} = \left[\begin{array}{c} h^2 \\ C_z^2 \end{array}\right] = \frac{1}{2-n}$$

(2)

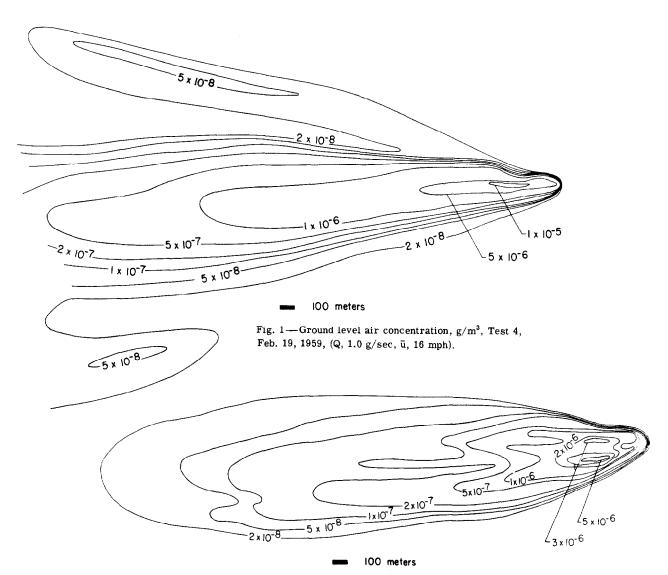


Fig. 2—Ground level air concentration,  $g/m^3$ , Test 6, Mar. 18, 1959,  $(Q, 0.79 \text{ g/sec}, \overline{u}, 22 \text{ mpn})$ .

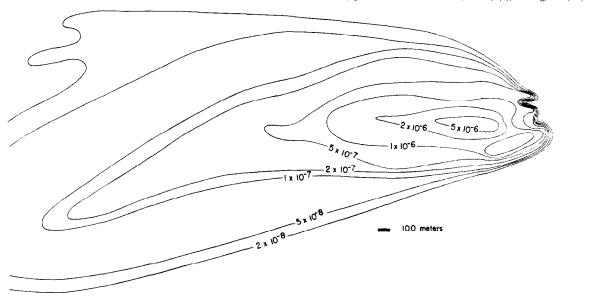


Fig. 3—Ground level air concentration, g/m³, Test 15, May 4, 1959, (Q, 0.88 g/sec,  $\overline{u}$ , 15 mph).

	100 B	×	1.1	M	1.3	1.5	1.4	1.9	1.9	M	2.5	1.0	1.6	1.4	1.7	1.5	2.3
	χ(c)/ χ (m)	2.8	2.8	0.8	14.2	2.8	3.0	<b>†*9</b>	3.2	4.1	3.8	2.5	9.9	1.6	1.8	3.0	4.3
	Computed X max (g/m <sup>3</sup> )	1.4x10-5	2.2x10-5	1.5x10-5	3.7x10-5	1.1x10 <sup>-5</sup>	1.2x10 <sup>-5</sup>	1.4x10-5	9-01x0-6	9-01x0-6	1.3x10 <sup>-5</sup>	1.7x10-5	9-01x6.7	1.0x10-5	1.3x10-5	7.8x10-6	1.3x10-5
	C 2 3 (m)	0.091	0.100	0.035	0.083	0.238	0.190	0.289	0.092	0.250	0.442	990.0	0.113	0.085	0.231	0.112	5 <del>1</del> 1/10
TESTS	$\binom{C_z^2}{(\mathbb{m}^n)}$	0.025	680.0	0.054	0.032	0.028	0.032	0.032	0.041	0.015	0.082	0.015	0,041	0.035	0.089	0.023	0.110
TABLE 2 DATA FROM URANINE DYE TESTS	ŭ <sub>1</sub> μο (m/sec)	5.7	6.5	0.7	8.9	2.6	7.8	7.2	10.0	7.7	6.7	8.1	11.3	0.6	6.5	10.6	6.5
DATA FROM	$\chi_{\rm max}$ (g/m <sup>3</sup> )	9-0x10-6	7.9x10-6	1.8x10-5	2.6x10 <sup>-6</sup>	9-01x0.4	9-01×0.4	2.2×10-6	2.8x10-6	2.2×10-6	3.4x10-6	9-01x19	1.2x10-6	6.1x10-6	7.4x10-6	2.6x10-6	3.0x10-6
	Assumed n	0.25	0.20	0.20	0.20	0.25	0.25	0.20	0.20	0.25	0.20	0.25	0.20	0.20	0.20	0.25	0.20
	dmax (meters)	700	300	700	530	650	009	560	024	046	315	046	09†	084	300	730	250
	(pes/8)	0.77	1.39	1.00	3.20	61.0	0.88	1.00	0.88	0.88	0.88	1.32	0.88	0.88	0.88	0.88	0.88
	Run	Q	က	†	2	9	7	80	6	91	11	12	13	14	15	16	17

Standard deviations of lateral wind direction fluctuation Standard deviations of vertical wind direction fluctuation (M indicates Missing) कि/न

The lateral diffusion coefficient Cy was determined from the following definition:

$$\frac{1}{y^2} = \frac{1}{2} c_y^2 \times (2-n)$$
 (3)

It is valid only if we assume that the variance of the dye on the arc chosen (in this case the arc closest to  $d_{\max}$ ) is in fact the same variance as Sutton uses in his definition.

The values of  ${\rm C_2}^2$  range from 0.015 to 0.11 meters  $^{\rm n}$ . The values were somewhat lower than expected but still in the range of values generally accepted. The low values may have been caused by the relatively high wind speeds associated with most of the tests which probably reduced the looping that would have been expected with the lapse rates realized.

The values of C range from 0.035 to 0.445 meters <sup>n</sup>. The values are somewhat larger than those used by many authors but it is felt that they are realistic when it is considered that no attempt was made, at this time, to remove bimodal tendencies or wind shear effect. The shear effect will be discussed later. These effects on the arcwise concentration distributions will obviously increase the lateral diffusion coefficient.

Sutton's diffusion model is based on a normal distribution, therefore, the normality of the concentration and wind data should be examined. Haugen [-9] has set up criteria for values of skewness and kurtosis in the study of effects of sampler spacing on concentration data. The values for determining if the distribution is normal are: for skewness  $B_1 \leq 0.10$ , and for kurtosis  $2.6 \leq B_2 \leq 3.4$ : borderline cases are classified from  $B_1 \leq 0.10$  and  $1.70 \leq B_2 \leq 4.20$ . Values for  $B_1$  and  $B_2$  for arcwise concentration distributions and the 140-foot level wind direction distribution are presented in Table 3. As indicated below in the table, only 17% of the cases of concentration data were normal. The addition of the borderline cases brings the total to 45% of all cases. The wind direction distribution at the height of the source is somewhat better, however, an inspection of the wind direction distribution at 10 feet indicates fewer cases of normal distribution than the concentration data. The kurtosis is about equally divided, in the concentration data, between platykurtic (curve too flat) and leptokurtic (curve too peaked). The leptokurtic cases were most frequently associated with skewed distributions.

The breakdown of the data at the end of the table indicates the number of normal cases of arcwise concentration distributions. There were 84 cases, of these 14 met the criteria mentioned above for normality and 24 additional cases were included under the definition of borderline normality. The remaining 46 cases did not meet the specified criteria for normality. The kurtosis of the curves is also indicated. The borderline cases are included since the interest here is only in whether the curve is platykurtic (flat) or leptokurtic (peaked). The curves were too flat in 28 cases and too peaked in 32 cases, therefore, only 24 cases were actually between the limits of kurtosis specified.

Figure 4 illustrates values for B<sub>2</sub> equal to 2, 3 and 4 so the reader may obtain a better understanding of the previous discussion. B<sub>2</sub> is equal to three for a normal distribution.

Figure 5 illustrates the variation of the cross plume standard deviation of concentration downwind from a continuous elevated point source. The tests were summarized according to the standard deviation of the 140-foot wind direction fluctuations. The range of the azimuth wind standard deviations is

TABLE 3  $\mbox{VALUES OF SKEWNESS (B$_1$) AND KURTOSIS (B$_2$) }$ 

TEST		2	3	4	5	6	7	8	9
ARC									
150	B <sub>1</sub>								N
	B <sub>2</sub>								N
300	B <sub>1</sub>	0.13	0.84	0.003	0.92	0.06	0.94	0.08	0.95
	B <sub>2</sub>	3•77	2.24	2.31	3.01	2.94	3.14	2.97	0.19
400	В	0.01	0.04	0.11	0.14	0.04	2.46	0.28	0.11
	B <sub>2</sub>	2.90	2.05	4.52	2.24	5.07	6.59	4.15	0.33
600	B <sub>1</sub>	0.00	0.01	0.67	0.08	0.04	0.28	0.09	0.01
	B <sub>2</sub>	2.93	1.96	4.21	2.69	2.11	3.42	1.95	2.49
1000	В	0.00	0.11	0.00	0.34	0.02	0.00	0.48	0.15
	B <sub>2</sub>	2.68	3.05	3.25	3.20	2.23	2.03	3.30	3.83
1800	B <sub>1</sub>	0.13	0.19	0.17	0.44	0.20	0.25	0.02	0.09
	B <sub>2</sub>	3.82	2.92	5.48	3.19	3•33	2.51	1.73	3.46
WIND	Bl	0.02	0.004	0.04	0.10	0.06	0.06	0.02	0.35
	B <sub>2</sub>	2.98	2.40	5.46	3•99	3.27	3.60	2.65	3.15

TEST		10	11	12	13	14	15	16	17
ARC									
150	Bl	0.038	1.51	N	0.13	N	0.95	N	2.18
	B <sub>2</sub>	3.09	6.07	N	4.03	N	3•74	N	8.37
300	B <sub>1</sub>	0.013	0.16	0.36	0.18	0.24	0.03	0.34	2.38
	B <sub>2</sub>	2.47	2.38	1.56	3.67	4.56	4.24	4.60	5.46
400	B <sub>l</sub>	0.05	0.46	0.19	0.69	4.34	0.32	0.03	4.17
	B <sub>2</sub>	2.79	3.10	3.11	7.12	8.27	4.58	4.15	7.88
600	B <sub>l</sub>	0.09	0 • 1474	0.03	0.01	0.18	0.04	0.10	2.71
	B <sub>2</sub>	1.94	4.45	1.89	1.84	3.65	2.63	3.88	6.38
1000	B <sub>l</sub>	0.17	0.10	0.06	M	0.00	0.13	0.01	1.16
	B <sub>2</sub>	2.41	2.17	2.48	M	2.76	10.4	2.30	5.65
1800	B <sub>1</sub>	0.26	0.04	0.10	0.07	0.00	0.19	0.00	2.63
	B <sub>2</sub>	2.85	1.96	3.21	2.59	2.01	4.98	2.25	8.60
WIND	В1	0.17	0.09	0.09	0.00	0.011	0.01	0.09	0.44
	B <sub>2</sub>	5.08	4.30	2.77	6.90	2.63	2.60	3•34	3.86
	COI	NCENTRATIO	ATAC V				WIND DA	ATA	
84 Cases 28 Platykurtic B <sub>2</sub> 2.6 16 Cases 1 Platykurtic B <sub>2</sub> 2.6 14 Normal 32 Leptokurtic B <sub>2</sub> 3.4 7 Normal 7 Leptokurtic B <sub>2</sub> 3.4 24 Borderline 3 Borderline									

<sup>(</sup>M indicates missing)

<sup>(</sup>N indicates no measurable concentration)

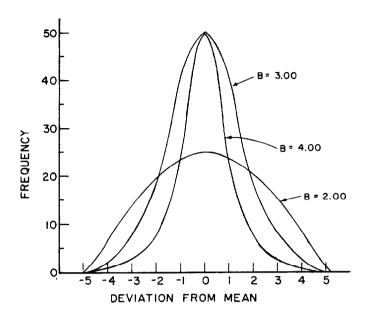


Fig. 4.—Curves illustrating kurtosis for values of  $\boldsymbol{B_2}$  indicated ( $\boldsymbol{B_2}$  equal to three is a normal curve).

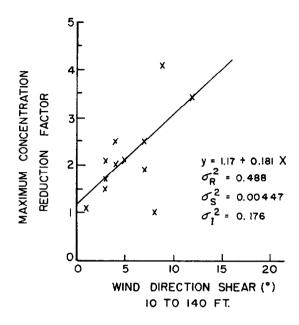


Fig. 6 - Mean wind shear effect on maximum concentration.

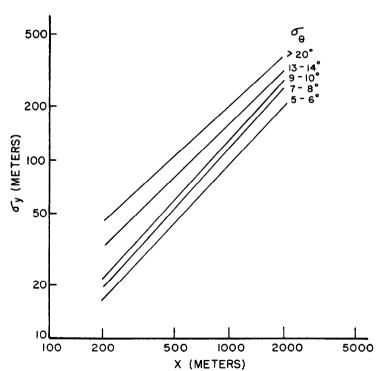


Fig. 5—Crossplume standard deviation of concentration downwind from a continuous elevated point source.

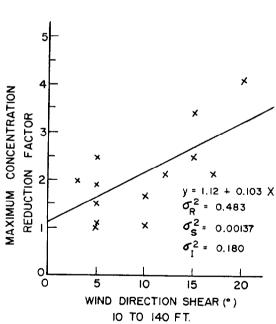


Fig. 7-Modal wind shear effect on maximum concentration.

indicated to the right of the lines in the figures. A recent paper by Cramer / 10 / using ground source data extrapolated to an elevated source indicates a slight convergence between near neutral and extremely unstable cases of crossplume standard deviation of concentration at distances beyond 2000 feet. There were insufficient data available to obtain any curvature to the lines in Figure 5 but there is an apparent tendency toward some convergence indicated at the extreme distances used in these tests. An analysis of the standard deviation in degrees of the arcwise concentration distribution indicates that the spread of the plume is not rectilinear, that is, the plume does not intercept the same number of degrees of arc at increasing distances. From this it is evident that the spread is a function of the distance frequently assumed to approximate some power law. This particular subject will be dealt with in another paper.

One of the most interesting results of these tests was the effect of vertical wind direction shear on the maximum ground level concentration. The computed maximum concentrations were consistently greater than those actually measured. Some of this may be attributed to the difference in sampling time between the three minute periods used to derive the equations by Sutton and the 30 minute sampling periods used in these tests and part of the difference may be attributed to the difference in the horizontal and vertical diffusion. For this study we have ignored the time difference factor and assumed that the vertical and horizontal diffusion are proportional to the vertical and horizontal deviations of a bivane. The maximum concentration equation was adjusted for the anisotropic turbulence in the following manner:

$$\chi_{max} = \frac{2Q}{e\pi \pi h^2} \frac{\sigma_{\bar{\epsilon}}}{\sigma_{\bar{\delta}}}$$
 (4)

where  $\sigma_{\epsilon}$  and  $\sigma_{\delta}$  are the standard deviations of the fluctuations of the vertical and horizontal wind direction traces. This factor was then removed from the ratio computed  $\chi_{\text{max}}/\text{measured}\,\chi_{\text{max}}$ . Both of these factors are listed in Table 2. The remaining reduction in the maximum concentration data was attributed to vertical wind direction shear causing the plume to be spread laterally more than would be expected in an atmosphere with a steady mean wind.

It was difficult to decide if the mean wind shear or the modal wind shear (i.e., the shear between peaks in the frequency distribution) were more important. Both cases were tested and are shown in Figures 6 and 7. The line of best fit was computed by the least squares method. The residual variances ( $\mathcal{T}_{R}^{-1}$ ) were computed to determine which represented the best method. Since the values were nearly identical the variances of the slope and intercept were then computed for each figure in a further effort to identify the best procedure. The variances of the intercepts ( $\mathcal{T}_{I}^{-1}$ ) are again nearly equal. The variances of the slopes ( $\mathcal{T}_{I}^{-1}$ ) indicate that the modal direction shear offers the best fit to the data. The differences are still so small that it would seem many more tests are required to determine which method should be used.

It should be noted that the samplers were at about three feet while the shear was only measured from 10 to 140 feet, so some error may have been introduced.

### Conclusions and Recommendations for Future Studies

Uranine dye stuff has been an excellent tracer material for the tests conducted to date. The particle size is in the aerosol range and analysis is rapid and uncomplicated. The values of the stability parameter n were

assigned but the values of diffusion coefficients have been calculated from the available data. These values were in accord with values generally used in atmospheric diffusion calculations. The values of the lateral diffusion coefficient (Cy) were somewhat larger than usually found and this may be attributed to two factors at least; 1) the meander of the plume becomes effective with sampling periods of thirty minutes creating wider plumes and 2) the vertical wind direction shear also creates a wider plume adding to the increase in lateral diffusion.

Future studies along this line should include samples in the vertical in order to obtain a complete budget of the dye. Even though particle size distributions have been determined and the aerosol range is realized, it would be best to determine the complete budget of material.

Plans for studies with the present data include testing various theoretical diffusion models and computing spectra of the horizontal and vertical wind fluctuations to determine the period and size of eddies most important to diffusion. The wind direction standard deviations are greater than those of the arcwise concentration distributions which indicates that a longer averaging period may be applicable rather than the five second means used.

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### Discussion

- C. I might give a few comments regarding the use of tracer techniques since our original paper, we have had considerable more experience with fluorescent dyes some of the problems you might get into fluorescent dyes cost about \$2 \$5 a pound. economical for studies lasting a few days which can use about 100 pounds a day if you have a study that goes on for weeks and months, chemical costs can be quite sizeable in long range diffusion studies. now in those cases we have used can use a few pounds a day and still get measurable quantities a few miles away and still get unknown quantities from the analytical procedure the filters experienced in our paper are not necessarily also the light fuel may not be pH control might be mentioned in trying to eliminate background. changing from pH 5 to pH 7 Filter leaching may be important. Reaction of background material dealing with extremely minute quantities also multiple fluorescent dyes may be used.
- Q. I would like to know whether the uranine or the other smokes or fumes can be used for predicting the concentrations of the bases the the noble gases. How confident are you?
- A. We have the uranine dye particulate size down to aerosol size, but it is still a particulate. I think we can make a fair approximation, but would hesitate to say that that actually duplicated the diffusion of the noble gases, because we are dealing with particulates rather than gas, but they would come very close.

### C. Lapple - Stanford Research

I would like to give a few comments regarding the uranine tracer type leak. Since our original paper, we have had considerable more experience with the use of fluorescent dyes and I would like to give a few comments, not any details, but just some of the problems you might get into. The fluorescent dyes generally cost about 25 to 45 a pound, so consequently if you have a study that lasts a few days you can afford to admit that at the rate of a couple of hundred pounds a day without incurring excessive chemical costs. If you have a study that may go for week or months then the chemical costs might become quite sizeable, as it might be in a long range diffusion tracer study. Now in most cases we have used ignition rates as low as a few pounds a day and still get measureable quantities several miles away from the admission point. However you do run into certain problems that have to be carefully considered. For example, in the analytical procedure the filters that are specified in our paper are not necessarily optimum, also the light source may not be optimum. These both may be influenced by the natural background of the materials. Ph control may be important in the analysis. For example, in the case of uraline you can get more than a ten-fold increase sensitivity by

changing a Ph from Ph 5 to Ph of 7. The analysis right in that critical region can be used in distilled water. Also the Ph control may be important in trying to eliminate background as an item. Filter leaching may be important. It may be difficult to leach. We believe that this is associated with reaction with background materials where you are dealing with these extremely small quantities. Also multiple fluorescent dyes can be used and analyzed simultaneously where you may want to trace from the missions of different points at the same time.

### Q. Newell - AEC Washington

I would like to ask a question as to whether the uranine or these other smokes are fumes can be used for predicting the concentrations of the gases - the noble gases, and how confident are you?

### A. Bowne

We have the uranine dyed particles size down to the aerosol range but it is still a particle. I think we could make the fare approximation to the noble gases but I would hestitate to say that they actually duplicate the diffusion of the noble gases because we are dealing with a particle rather than a gas. They would come fairly close because they are a fairly small particle.

### C. Silverman:

Not being inhibited by being a meteorologist I would go at that question to say that it would probably behave very much like it because the studies of Barren(?) and Johnstone indicated that under five microns you can consider the behavior practically as that of a gas - if you neglect depletion rates.

### ESTIMATES OF ACCUMULATED EXPOSURES AND ENVIRONMENTAL BUILD-UP OF RADIOACTIVITY

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### ABS TRACT

A simple method for estimating long-term effluent concentration and deposition values based on Sutton's and Chamberlain's equations is presented. Calculations of concentration and deposition are straight forward, and become especially convenient if averaged wind data are employed.

ESTIMATES OF ACCUMULATED EXPOSURES AND ENVIRONMENTAL BUILD-UP OF RADIOACTIVITY

An approach to long-range effluent concentrations, similar to that of Meade and Pasquill /1/, was attempted at Oak Ridge. Hampered by bombtest fallout and instrument variation, experimental verification as striking as Meade and Pasquill's results with SO<sub>2</sub> was not obtained. More elaborate and time consuming, the methods outlined below are, nevertheless, easily applied, and utilize existing wind information to the fullest extent practicable.

Meade and Pasquill, as well as earlier works by Holland 27, and Lowry 37, used the mean wind speed of each sector (direction) of interest. In our Oak Ridge studies we are going one step further by using the mean wind speed of each of six speed groups of every sector, giving a finer breakdown of wind behavior. We also are using the sixteen point direction scale, common to most wind rose publications in the U.S. Two sets of "Work Sheets" are prepared to facilitate computation, one for average air concentrations, the other for deposition calculations. Either can be used directly by untrained help, or can be modified to fit specific situations by anyone familiar with a slide rule and equations (4a), and (7a), below.

The Sutton formula for concentration at ground level from a steady, continuously emitting, elevated source is:

$$\chi = \frac{2Q_o}{\pi (2\chi^2 - nu)} \exp \left[ -\left(\frac{h^2 + y^2}{c^2\chi^2 - h}\right) \right]$$

(1)

where  $\chi$  = concentration (at ground level); parts/meter<sup>3</sup> = emission rate; parts/second  $\frac{n}{3}$ 

= diffusion coefficients; meters 2

n = stability parameters; non-dimensional

= distance downwind from source; meters

= distance crosswind from plume centerline; meters

= average wind speed; meters/second

= height of source; meters

and isotropic dispersion is assumed [4.7.

The quantities in equation (1) that are assumed to vary significantly during a relatively long period are C, n, and  $\pi$ .  $Q_O$  and h are assumed constant over a year's time. It is not unreasonable to assume that, on a seasonal or annual basis, the daytime n-value would approach the "neutral" stability level (n = .25), and the nighttime n-value would approach the "moderate inversion" value (n = .33). Wind speed values and the n-values then determine the remaining variable, C, as a function of a stack height. A nomogram convenient for determining C appears in reference 27.

Wind direction data are often based on 16 compass points, and this number will be assumed here. These 16 "points", however, are in reality 22.50 sectors. At five miles distance downwind, direct application of equation (1) might lead to an error of 5 orders of magnitude for any given point unless the average wind direction is known exactly. Over short distances and times, the error involved is not serious. Over a year's time, however, we cannot assign one direction to "north", for instance; "north" must rather be considered as an entire sector of 22.50. This is achieved mathematically by integrating eq. (1) with respect to y and then distributing the result uniformly along the entire arc. Thus,

$$\frac{16}{2\pi\chi} \int_{-\infty}^{\infty} \chi dy = \frac{16}{2\pi\chi} \int_{-\infty}^{\infty} \frac{2Q_0}{\pi c^2 \bar{u} \chi^{2-n}} \exp\left[-\left(\frac{h^2 + y^2}{c^2 \chi^{2-n}}\right)\right] dy$$

(2)

$$= \frac{/6Q_0}{\pi^{5/2} \text{ ac } x^{\frac{4-n}{2}}} \exp \left[\frac{h^2}{c^2 x^{2-n}}\right]$$

(3)

Equation (3) is modified to account for the annual wind speed and direction point frequency distribution (wind rose) in the following way. For a fixed n,

$$\overline{\chi}_{D} = \sum_{i=1}^{R} F_{i} \frac{16Q_{o}}{\pi^{\frac{4}{2}} c_{i} \times \frac{4-n}{2} \overline{u}_{i}} \exp \left[ \frac{h^{2}}{c_{i}^{2} \times 2-n} \right]$$
(4)

where ui = average speed for wind speed group "i"

 $F_1$  = fraction of total time wind is from direction D, and speed group i.

C = diffusion coefficient determined from n, Ti, h.

D = annual average concentration along a 22.50 arc, at distance x, in direction D.

R = index denoting the number of wind speed groups in the annual distribution.

To facilitate hand compution, equation (4) may be written as,

$$\overline{\chi}_{D} = \sum_{i=1}^{R} \text{Fi } \beta$$
where  $\beta = \frac{\frac{6Q_{o}}{\pi^{3/2} \text{ c.i.} \times \frac{4-n}{2} \text{ u.i.}}}{\text{exp}\left[-\frac{h^{2}}{c^{2} \times 2-n}\right]}$ 
(ha)

Sixteen work sheets, one for each wind direction, are convenient, (32 work sheets if both night and day are computed). Figure (1) is a sample calculation sheet.

Figure (1) lists "Day" and "Night" conditions. When available, wind roses using "lapse" (for day) and "inversion" (for night) conditions are used in preference to merely daytime vs. nighttime wind roses. The number of hours of inversion varies with the location of the site. At Oak Ridge, we find the hours of inversion distributed as in Table I.

TABLE I
Average Daily Hours of Inversion by Season (1955-1958)

	Winter	Spring	Summer	Fall
TSF	10.1	10.1	12.7	12.7
MV	11.1	11.1	11.1	12.7
X-10	8.1	9.2	9•3	9.8

Where TSF = Tower Shielding Facility, 5-300 ft. above a wooded ridge top, (500 ft. from the valley "floor").

MV = Melton Valley, 5-60 ft. from the "floor" of a wooded valley.

X-10 = Oak Ridge National Laboratory site, 5-135 ft. atop a small ridge overlooking an industrial type valley, 90 ft. below.

Calculations for the Cak Ridge dispersion patterns are based on the TSF data, since the height of the stack plus plume rise through bouyancy would approximate the TSF height.

 $X = B \times Fi = parts/meter^3$ 

										2 24 hr							
		· · · · · · · · · · · · · · · · · · ·							ı	2 Night							
	25+	0	8.6 x 10°9	5,5 x 10-8	7,2	6,3	. 1.7	2,7		25+						1	
	20-24	0	1.8 I 10-8	7.8 "	. 7.6	7.8 "	" 6 <b>.</b> 7	3,3 "		20-24							
n=.33	15-19	٥	2.9 x 10-8		1,3	1.0	6.3 x 10-8	4.2	iΞ	15–19					}		
NIGHT n=.33 B	10-14	0	4.0 1 10-8	1,6 x 10-7 1,2 x 10-7	1,7	1,4	8.8 I 10-8	5.8 "	B x Fi	10-14							
	5-9	0		3,3 "	=	2,5 "	1,4	9.3 X 10-8		5-9							
	u=1-4mph	1,4 x 10 <sup>-10</sup>	1.0 x 10-6 1.3 x 10-7	1,2 "	9.9 x 10-7 3.2	. 7.9	3.5 "	2,2 "		u=[1-4mph							
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	25+	1,3 I 10-7	2,1 **	1.1	5.9 I 10-8	3.2 "	1.5	9.0 x 10-9		25+ \\\\ \\\ \\\\\\\\\\\\\\\\\\\\\\\\\\\							
		2.0 I 10-7 1, 3 I 10-7				•	1,6 * 1,5 *	1,1 " 9,0 x 10-9		25+							
1=.25 3	20-24		* 2,1	. 1.1	5,9 I 10-8	* 3,2 *	•	1,1	ί <u>τ</u>	-19 20-24 25+							
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Day n=.25 B	20-24		n 3,3 n 2,6 w 2,1	. 1,7 . 1,3 . 1,1 .	* 9.0 x 10-6 7,1 x 10-6 5,9 x 10-8	4,8 " 3,8 " 3,2 "	. 2,3 . 1,6 .	1,8 1,3 1,1 1	B×Fi	15-19 20-24 25+							
Day n=.25 B	10-14 15-19 20-24	4.1 x 10-6 1.0 x 10-6 4.7 x 10-7 3.0 x 10-7 2.0 x 10-7 1.3 x 10-7	4,6 " 3,3 " 2,6 " 2,1	. 2.3 . 1.7 . 1.3 . 1.1 .	1,2 " 9,0 x 10-6 7,1 x 10-6 5,9 x 10-8	. 6,5 X 10-8 4,8 " 3,8 " 3,2 "	3,1 " 2,3 " 1,8 "	1,8 " 1,3 " 1,1 "	ïΞ×Β	10-14 15-19 20-24 25+							

Fig. 1—Sample work sheet for determining average annual effluent pattern, at surface; source height, h=100 meters; source strength  $Q_0 = 1$  part/sec.

#### Deposition

Once the average air concentrations are calculated, the average deposition (based on Chamberlain's method) [5\_7, [6\_7, may be computed. Deposition may be said to occur as,

$$\phi = \chi v_{g} \tag{5}$$

where  $\emptyset$  = deposition, parts/meter<sup>2</sup> second

X = concentration, parts/meter<sup>3</sup>
Vg = Velocity of deposition, meters/second
 has already been calculated from the effluent pattern worksheets for various distances downwind. As deposition occurs, however, the effective source is diminished, therefore X should be multiplied by a "diminution factor", Q/Qo, which is supplied in the deposition worksheet, figure 2.

Figure 2 could have been adjusted to read  $(Q/Q_0)V_g$ , instead of  $Q/Q_0$ , but  $Q/Q_0$  required adjustment when  $V_g \neq .04$  meters/second. Should  $V_g = K$  meters/second, the deposition worksheet may be modified by raising  $Q/Q_0$ to K/.04 power. Thus for any  $V_g = K$ ,

$$\frac{Q_i}{Q_o} = \frac{Q}{Q_o}$$
(6)

Any change in effective source height, however, will alter  $Q/Q_0$ in a manner not easily calculated. To recalculate deposition values for  $h \neq 100$  meters, the formula;

$$\ln \frac{Q_2}{Q_0} = \frac{C_{21} u_1 v_{g2}}{C_{22} u_2 v_{g1}} \ln \frac{Q_1}{Q_0}$$
 (7)

and figures 3 and 4 may be used, [6]7.

= desired depletion factor

 $\frac{Q_1}{Q_2}$  depletion factor from figure 3 or 4

 $^{C}zl = diffusion coefficient from figure 3 or 4$ 

ul = wind speed from figure 3 or 4

 $V_{gl}$  = deposition velocity from figure 3 or 4

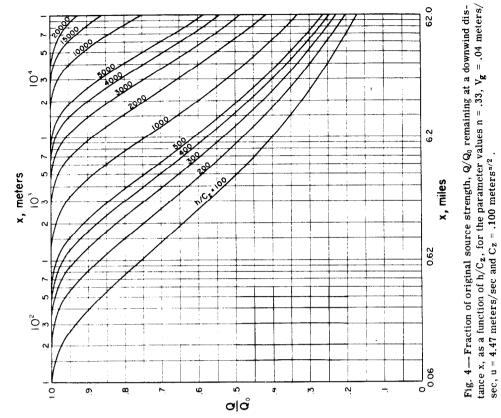
 $C_{22}$  = altered diffusion coefficient

un = altered wind speed

Vgl = altered deposition velocity

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Direction 18											ENight   2 24 hr.						
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je Se	NIGHT n=.33 Q/Qo	10-14	00%	7.00	96.	88.	97.	. 58	34.	0/0° × X × Vq	10-14						
parts meter <sup>2</sup> × sec.		5-9	7.00	00'/	76.	41.	09:	.38	97'	9	5-9						
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		ı — ı							20								20

Fig. 2—Sample work sheet for determining average rate of deposition of material at surface; source height, h = 100 meters, and velocity of deposition,  $V_g = .04$  meters/sec.



3

0

x, meters

30.5

0

Ø,

80

3000

Fig. 3—Fraction of original source strength,  $Q/Q_0$ , remaining at a downwind distance x, as a function of  $h/C_z$ , with parameter values n = .25,  $V_g$  = .04 meters/sec, u = 4.47 meters/sec and  $C_z$  = .100 meters<sup>n/2</sup>.

62.0

2.9

x, miles

0.62

0.06

Ņ

w

Ŋ

ø

Since  $C_{2l}$ ,  $u_l$  and  $V_{gl}$  are all constant, equation (7) becomes

$$ln \frac{Q_2}{Q_0} = \frac{11.18 \text{ Vg}_2}{\text{C} = 2 \text{ U}_2} ln \frac{Q_2}{Q_0}$$
(7a)

The deposition work sheet may thereby be altered to fit any given set of parameters.

# Plotting the Results

Figures 5 and 6 are representative plots of average daytime air concentrations (at the surface) and average daily deposition rates. Strictly speaking, the values should have been plotted as a series of discontinuous 22.5° arcs. For convenience, however, the calculated values were plotted at the center of each representative arc and isopleths smoothed to fit.

Deposition equations 5-7 are restricted to values of Vg << u.
Thus, the deposition during "calm" cases, (or more to the point, at wind speeds less than detectable by the anemometer employed) must be treated in a different manner. An obvious method is;

$$\phi_c = \frac{F_c Q_o}{\pi G^2} \tag{8}$$

where Øc = deposition during "calm" cases.

G = radius equal to the magnitude of the anemometer's starting speed. Equation 8 is not entirely satisfactory, but it serves as a basis for subjective analysis when the entire deposition pattern is drawn.

After plotting the average concentration and deposition values by use of figures 1 and 2, similar plots were made using the only mean wind speed of each direction multiplied by frequency of winds in the sector of interest. The results of both concentration and deposition values using the mean wind of each direction were very close to those obtained by using the entire wind distribution. It appears that computing effluent behavior by use of the mean wind is probably a very good approximation to the longer methods otulined above. Nevertheless, considering the applications of effluent predictions, one to two hours more effort seems worthwhile.

#### Further Considerations

An interesting and possibly quite useful extension to multiple point and area sources of the foregoing technique follows from a suggestion made by Gifford /7/. If there are, instead of a single point source, any number of arbitrarily located point (or small area) sources of arbitrary strength (but all at equal heights), the resultant annual concentration or deposition value at any ground-level point may be calculated easily by reversing the role of source and receptor in Figures 3 through 8. One would simply rotate the appropriate figure through 180° (i.e. turn it bottom side up), and orient the center point at the ground-level receptor location. If then the actual source locations be plotted on this figure, and for each such point the product of the concentration (or deposition) isopleth and the corres-

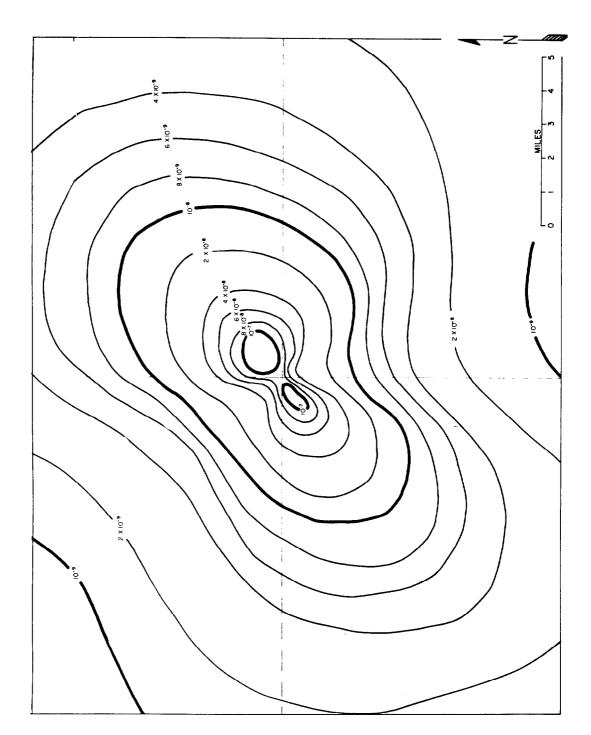


Fig. 5—Average daytime surface concentrations of effluent, based on yearly wind data; source height h=100 meters; source strength,  $Q_0=1$  part/sec, n=.25.

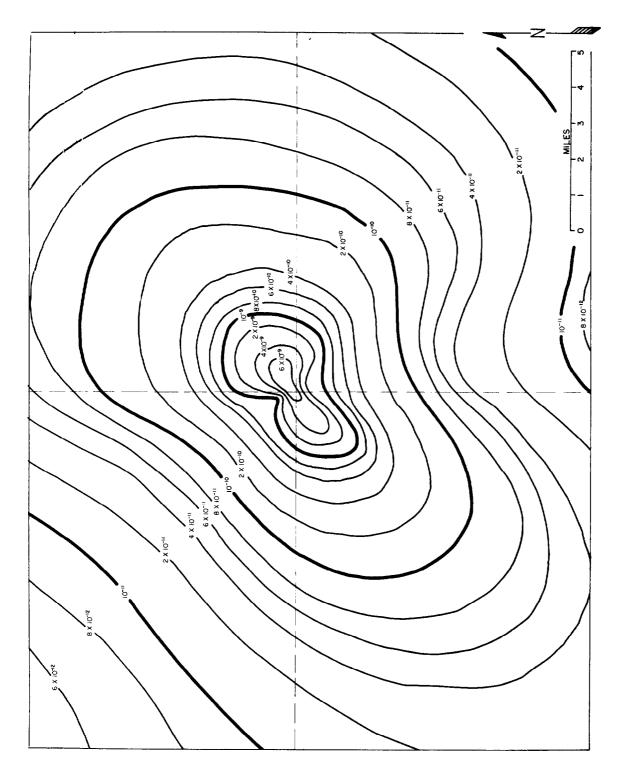


Fig. 6 —Average daytime deposition rates, based on yearly wind data; source height, h = 100 meters,  $V_g$  = .04 meters/sec,  $Q_0$  = 1 part/sec.

ponding source strength be formed, then the total annual concentration or deposition value at the receptor (center) point is given by the sum of these products. This process can be repeated for various receptor locations, thus providing annual distributions for multiple or area sources.

### Acknowledgement

The author wishes to thank Mr. Paul Rhodes of the Oak Ridge Weather Bureau Office for preparing the figures in this paper.

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# ANL AIR CLEANING RESUME

#### C. L. CHEEVER

Argonne National Laboratory, Argonne, Illinois

First, I will make a few comments on the various types of air cleaning equipment in use at Argonne, and then briefly discuss air cleaning plans for our new Fuels Technology Center.

High efficiency filters, in conjunction with prefilters, carry the major portion of the exhaust air cleaning load at the Laboratory. Generally, there are about six individually mounted filters per exhaust plenum. Most have fiberglass media and aluminum separators, although filters with asbestos media and some with asbestos separators are used for special requirements. Filters with paper separators have not been entirely replaced, but they are fading fast as all replacements are with the non-combustible separators. Recently, some filters with honeycomb construction have been purchased.

Our experience with high efficiency filters has been generally good, with a few exceptions that can be attributed to excessive insult to the filters.

In one case, glass media filters were installed in an exhaust system where there was the possibility of release of hydrogen fluoride. Evidently, hydrogen fluoride was released to two of the high efficiency filters as the filter media disappeared. This was easy to pick up on a check of the pressure drop across the aluminum separators.

In another instance, considerable laboratory acid fuming was carried out without an acid scrubber in the exhaust system. The media in the high efficiency final filter (with paper separators) dropped into the lower plenum.

A Millipore filter sample was run continuously at 1 cfm for 35 days on the exhaust side of a high efficiency filter serving a laboratory hood exhaust in our Chemistry Building. No detectable weight gain or reduction in flow rate was indicated. The Millipore filter had a gray discoloration, but there was no particulate loading that could be removed from it.

Prefilters are American Air Filter Company Type H and Type HX with fiberglass media. We find that the prefilters do a pretty good job of protecting the downstream ductwork against particulate contamination and in reducing the amount of particulates getting to the high efficiency filters.

The collectors used to clean the exhaust from operations in our Special Materials machine shop and foundry are a series combination of Type N Rotoclone, Electrocell Electrostatic Precipitator and Airmat paper filters. Efficiencies have ranged all the way from 60% to 98.8% for uranium and 95% to 99.9% for beryllium.

The cloth bag collector used for collection of considerable quantities of dust from our graphite machining room has proved satisfactory.

Building supply air filters are Type PL-24 with either Airmat paper or fiberglass media. Our tests have indicated average weight efficiencies for the two filter banks of 60-odd per cent against atmospheric dust.

The Venturi scrubbers for cleaning the exhaust from two sodium burn disposal booths do a good job. The problem is in limiting sodium fires to the burn booths.

Two large acid scrubber units with up to 6000 cubic feet per mimute capacity for uranium fluoride pilot plant cell effluent have three stages of nozzles spraying potassium hydroxide solution into Schutte and Koerting throats. Early tests on our first scrubber indicated efficiencies of 98% to 100% against hydrogen fluoride. In operation, the highest fluoride concentrations found during discharge stack sampling has been 2.24 micrograms per cubic meter.

Type D Rotoclones are used for scrubbing the effluent from acid pickling tanks in our Special Materials foundry.

Our acid fume scrubbers for laboratory fume hoods are small (150 cfm) units utilizing a spray wetted fiberglass pad of 100 to 200 micron diameter fibers, and identical dry fiberglass pad, and a high efficiency final filter. The high efficiency particulate filter is needed as acid particles penetrate the scrubber stage. Collection efficiencies are greater than 99%.

Our new Fuels Technology Center, which is presently under construction, will have a variety of air cleaning problems. This facility will be devoted to carrying out research projects on reactor fuel element development, and the information gained will be used in production at the Fuels Fabrication Facility.

Some 456,000 cfm will be exhausted from the building, and this air will be replaced with high velocity supply air (3500 to 4000 fpm) filtered with American Air Filter rollomatic roughing filters, and secondary Type PL-24 fiberglass media filters. The general philosophy of the building is to keep each laboratory as a separate unit so that only one laboratory would have to be incapacitated in the event of a spill or other release of activity. Holes for utility pipes are to be caulked and doors and louvers could be taped to seal off the room. Recirculation duct connections will be installed in the corridor wall so that if future need arises a portable high efficiency filter unit could be attached to recirculate and clean the room air.

High efficiency filters for the room exhaust will be installed within the room near ceiling height. The plutonium gloveboxes will have a nitrogen atmosphere with 0.2 to 0.5% oxygen content in a recirculation system, with high efficiency filters at the glovebox entry and outlet and at the entry to the oxygen-hydrogen catalytic combiner units. Every part of the system, with up to 20 boxes per system, will be under 0.5 or more inches of water negative pressure with respect to the room.

A plastic pouch thermal seal filter change technique will be used to prevent the header filters from becoming open to the building atmosphere.

Work is now in progress to fabricate and test an acid fume scrubber to be used for some of the acid fuming in the chemical laboratories. The scrubber will be constructed of fiberglass impregnated with Hetron 92 plastic containing 5% antimony trioxide. The exhaust air from acid fuming beakers will pass through a sprayed fiberglass pad, then through a dry fiberglass pad mist eliminator, and finally through a cartridge packed successively with 200, 25, 1, and 0.5 micron glass fibers. This cartridge can be back-flushed to remove collected acid. Dr. L. Silverman, Harvard University, suggested design features of this unit.

Exhaust air from the Metallurgy machine shop, foundry, and ceramics operations will be filtered with reverse jet cloth bag collectors. In the case of the ceramics toxic materials laboratory, where fine beryllium powders will be used, these collectors will be backed up with high efficiency filters. Efficiency is specified at 99.95% vs. 1 micron spheres.

Gas absorbing units for pickling, plating and other operations in the shop and foundry area will have wet and dry stages of dynel fibers and the interior will be coated with a minimum of 50 mil thick polyvinyl chloride.

Preoperational efficiency tests will be run on the air cleaning equipment for this building, with occupancy scheduled to start after the first of the year.

# SUMMARY OF AIR CLEANING ACTIVITIES AT ATOMICS INTERNATIONAL

E. C. HICKEY

Atomics International, Canoga Park, Calif.

At Atomics International, a Division of North American Aviation Company, activities are conducted primarily at two general site locations: (1) reactor, hot cell and critical assembly operations at the field site located in the Santa Susana Mountains and (2) research and development and fuel element fabrication operations at facilities located in Canoga Park section of the San Fernando Valley approximately 15 miles from the field site. Since a specific discussion of existing and proposed air cleaning installations relating to reactors, hot cells and critical facilities will be presented by A. R. Piccot of the Analytical Engineering Department at Atomics International, it would appear appropriate to confine these remarks to a brief discussion of current air cleaning activities as they relate to fuel element development and fabrication as well as basic research and development.

During the past year, considerable effort has been expended in the planning, design and construction of new facilities to meet the continually increasing demand for space to support R and D activities in the San Fernando Valley. Recently, construction was completed on a new building which will be devoted principally to fuel element development and fabrication. Construction is also underway at present on three (3) additional buildings which will complete a four (4) building complex. One of the latter buildings will be utilized exclusively for R and D laboratory work.

Due to the proximity and density of residential dwellings to the facilities in the San Fernando Valley, the basic criteria relating to the discharge of airborne radioactivity from these facilities can be summarized as follows:

Operation or processes that produce, generate or disperse radioactive gases or particulates are provided with adequate means for the collection and removal of such contaminants from the air prior to discharge to the atmosphere. It is required that the most efficient and practical system of air cleaning be provided to reduce activity level to a minimum and

at all times sufficient to maintain activity below prescribed tolerance levels.

Collection of particulates is usually accomplished by the use of 2 inch thick non-combustible fiberglass prefilters and non-combustible high efficiency final filters. In many instances, however, pre-cleaning of the exhaust air prior to final cleaning is provided at the point of dust generation particularly in powder handling and machining operations. Reverse-jet filters are also being employed in this capacity for such operations as the machining of beryllium and contaminated graphite dies. Gas collection and removal is provided in all cases at the point of generation. In most instances the gas is collected in hold tanks for controlled discharge after decay. When direct discharge of gas to a hold tank is not possible, then methods such as freeze-out, adsorption and absorption are utilized depending upon the nature of the material.

The recently activated fuel element development and fabrication facility where SRE, OMRE and other special fuel elements are fabricated is provided with four (4) air handling systems and the general room and local exhaust air requiring cleaning amounts to approximately 90,000 cubic feet per minute. Operations carried on within this area include: vacuum casting operations, heat treating, slug loading, machining, assembly, testing and all phases of metallographic testing (hot grinding, hot polishing, microscopic examination, corrosion testing). Provisions will also be made at this facility to fabricate the fuel elements for the Hallam and Piqua Power Reactors. For the R & D laboratory building presently under construction, seven (7) air handling systems will be installed with a total general room and local exhaust air cleaning requirement of greater than 100,000 cubic feet per minute. Operations to be carried on within this facility include: High and low level analytical and separations chemistry, radiometric measurements and analysis, high temperature ceramic studies, organic reactor material studies, electronic and standards laboratories, R & D machine shops, a laboratory reactor and numerous other activities related to the various applications of nuclear materials. These air handlings and high efficiency air cleaning systems represent a sizeable capital investment and maintenance expenditure in order to satisfy the fundamental criteria relating to the discharge of airborne radioactivity from these facilities. The location, nature and extent of operations, however, justify the installation of adequate air cleaning systems.

In order to continually evaluate the effectiveness or integrity of any air cleaning program, stack sampling and environmental monitoring programs are utilized. Stack sampling is used on the present major exhaust systems and will be incorporated into the new systems as they become activated. Environmental monitoring which is considered an important adjunct to any air cleaning program is utilized to assay both airborne particulate activity as well as surface contamination or fall-out resulting from possible plant discharges or nuclear detonations. An extensive environmental monitoring program is in effect at Atomics International which includes constant air monitoring at all facility locations, the routine collection and analysis of soil and vegetation samples from forty-three (43) stations covering a 15 mile radius from the field site at Santa Susana. These results continue to show that no significant contamination has resulted in the environs due to the operations

carried on within the area during the past 5 years. Meteorological data are also collected at Santa Susana which includes recorded wind speed and direction as well as temperature and relative humidity. These data supply necessary information which can be used for hazard evaluation studies as well as for emergency plans.

# SUMMARY OF AIR-CLEANING ACTIVITIES\*

#### MYRON D. THAXTER

Lawrence Radiation Laboratory, University of California, Berkeley, Calif.

#### ABSTRACT

The air-cleaning policy of the Lawrence Radiation Laboratory includes studies for improvement of methods and equipment. Among new developments and applications are sampling equipment and scrubbing or decontamination assemblies; further investigations are needed.

### General Statement

Our air cleaning policy is directed towards (a) improving the quality of air-cleaning equipment and practices, (b) reducing the contamination of air wherever processes and established routines permit, and (c) reducing the quantity of air exposed to contamination. The maintenance and enhancement of air-sampling equipment, methods, and data processing (including the interpretive reporting phase) continues as an essential portion of our air-cleaning program.

## Improvements in Methods

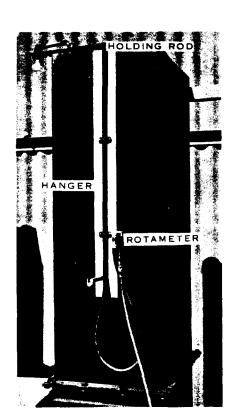
- 1. Lawrence Radiation Laboratory, Berkeley, has substantially abandoned the use of fume-hood exhaust filters. Over many years the amounts of radioisotopes worked up in hoods had been decreased (as the use of total enclosures increased) to the point where hood filters were functioning as rather poor stack samplers. When adequate stack-sampling equipment became available the filters were abandoned. The sampling frequency is now weekly, and evaluation criteria shall be according to 10 CFR 20.
- 2. Heavy-element slug processing became 100% functional under our so-called 'low leak" air-handling system. As a direct consequence, the stack samples for the first time were free of any detectable alpha activity.

Paper presented by H. Jorden

3. The so-called closed dissolver prototype, referred to at the 1957 seminar, has since been used on multicurie runs with quantitative capture of fission gases. No operating difficulties are experienced, and in the collection process the gases are automatically packaged and ready for burial.

## Development of Air-Handling Equipment

1. A hood-exhaust sampling assembly (Figs. 1, 2) has been devised, tested, and installed. About 3 months' data are at hand. Replicate samples show about a 70% agreement with one another on samples of 1 week's duration. Relatively minor mechanical problems awaiting solution are (a) pump shutdown and (b) rubber deterioration, in a few instances, on the sampler head assembly.



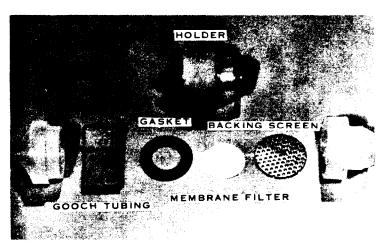


Fig. 2 — Stack sampler (exploded view).

Fig. 1 — Stack sampler assembly.

2. A general-purpose refrigerated scrubber and air-conditioning assembly for treating either recirculated or once-through process gases at a maximum rate of 18 cfm (Fig. 3) has been developed through a design study based on 8 years' experience at two sites. Several of these units have given successful service in high-level general radio-chemistry. Experience shows that, through operator negligence, the Venturi throat may become plugged with crystalline deposits. A method is needed for removing these crystals from "hot" apparatus.

Exploratory data on aerosol and gas-removal efficiency during a recent high-level run indicated from 40% to 96% per pass, depending upon the isotope measured. This variation was not unexpected because of the wide range of solubilities, vapor pressures, etc., in a heterogeneous mixture of fission products and transplutonic compounds.

3. An air-sample alpha and beta-gamma counter with auto-matic-printout--for 4×9-inch filter paper samples--has been devised

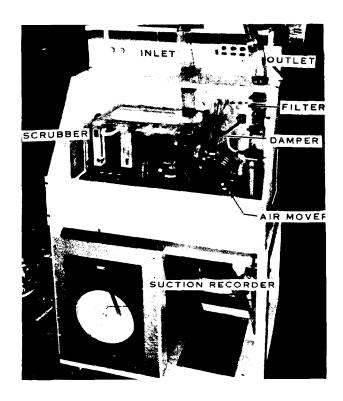


Fig. 3—Scrubber box assembly.

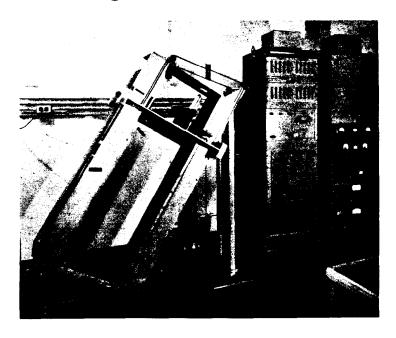


Fig. 4—Automatic filter-paper counter.

and substantially debugged, and is turning out routine data (Fig. 4). It can handle 85 samples per loading. A more detailed paper is in manuscript (M. D. Thaxter and T. G. Taussig, Automatic Filter Paper Reader, UCRL-8701, September 1959).

- 4. Modifications were made on a fail-safe self-checking circuit for box exhaust manifolds to employ commercially available components. We believe we now have a dependable assembly based on more than 2 years' use and development.
- 5. A prototype high-speed centrifugal evaporator or so-called "spin-dry" equipment shows promise; early runs indicate decontamination factors greater than 10<sup>5</sup>. It is expected that development will proceed to both macro and semimicro hardware.
- 6. Safe equipment and methods for taking 15-ml process gas samples in 1/4-mil Mylar bags were developed and employed (Fig. 5). The technique permits detection of weak beta and alpha radiations and performance of pulse-height analyses on gamma emitters.

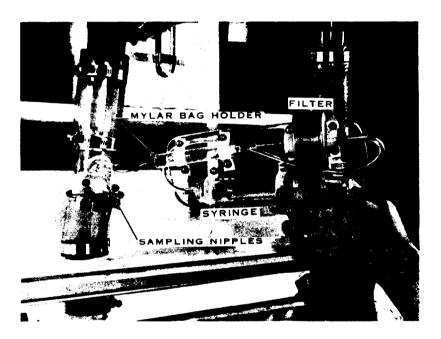


Fig. 5 - Air-grab sampler assembly.

#### Investigations Needed or in Progress on Methods and Equipment

- 1. Radioactivity is found in the air near operating accelerators. A portion is airborne. The extent, nature, method of production, and evaluation of hazard (if any) of this airborne fraction is under investigation. At least a portion of the activity is collectable on dust filters and emits beta particles. Half lives ranging from  $7\frac{1}{2}$  seconds to 40 hours have been observed.
- 2. Revelation of mechanical defects in high-efficiency filters (CWS 6 and AEC type) by visual inspection points up a need for technique and equipment to evaluate filter efficiency upon receipt. This is particularly important since only a very small part of the filter is visible to inspection.
- 3. Prompt detection in air of alpha emitters other than radon-thoron daughters is badly needed, and we continue to plead for research and development leading to a dependable and economical method.

4. Methods for safe, rapid evaporation of radioactive solutions, in addition to the "spin-dry" technique already mentioned, are the subjects of studies planned at this Laboratory.

# Discussion

- Q. (W. B. Harris, NYOO) About the hopper for collecting materials, I heard something about having a level indicator in the **bin** is this adequate protection against a critical amount in the hopper?
- A. It should never accumulate to that point at all. If it does, there is an alarm (which we hope will never go off).

# SPEECH DELIVERED TO 6TH AEC AIR CLEANING SEMINAR AT IDAHO FALLS, IDAHO, ON JULY 7, 1959

R. J. WALKER

Dow Chemical Co., Rocky Flats Plant, Denver, Colo.

In reviewing the radioactive air filtration problems at the Rocky Flats Plant in Colorado for this group at Boston two years ago, I stressed the fact that we had experienced very commendable service (in both efficiency and cost) from our cellulose-asbestos final air filters. I expressed our concern over the danger of fire in these banks but I also expressed some misgivings we had concerning the new, all-glass, fire resistant filters being proposed by one of the filter manufacturers. Since we, as well as many of you, were living on "borrowed time", so to speak, with nearly 4000 of the old "firebug" filters in place, I was especially concerned that all due emphasis and effort be placed on getting a suitable "standard" evolved for the new fire resistant filters so that we could all proceed with our mass replacements without jumping from the frying pan into the fire. This is probably an unfortunate choice of words which could be better expressed by saying we surely didn't want to jump into such a major program and find we bet on the wrong horse. My sole point in the talk was to press for a solution to this problem and that if furthering this end were our only accomplishment at the meeting that all of our time would have indeed been well spent.

Very unfortunately, time ran out on us. As most of you are well aware, we lost one of our major banks of these filters in a fire just three months after that Boston meeting. Luck, nature, design, and super efforts on the part of personnel involved all helped to minimize what could have been catastrophic consequences. However, ready or not, there was no more time to wait for the solution to the problems of the new, all-glass, fire resistant filters. Hanford had just ordered the first major batch of these new units. By utilizing filters intended for them and by working all three manufacturers overtime, we were able to have this bank back in operation in about two weeks.

However, since then, some of our worst misgivings about the new units have been realized. First, our costs have been staggering for numerous reasons. This is not just sour grapes from an enemy of the "quality at any cost" school. The first cost of the first hurried units was nearly 50 percent over the old-type units. In the first four months, 10 percent of the filters in the bank had holes and had to be patched. In the first six months, we also

had to completely replace another 10% of the bank. When the whole filter core is pulled out onto the floor, you have little choice. The man-hours needed to check, search for, and maintain such a bank become costly in dollars and rob important production functions. As a result, the whole bank was replaced with a new glass-asbestos type media filter in a year (up to being burned, the old bank had lasted five years). So far, the new units are doing a good job with much less effort on our part than the all-glass units. They do, however, require more attention (checking and patching) than the original bank.

The pressure of the crying need for the best in fire resistant final air filters has brought forth numerous variations and new designs. This, coupled with what seems to be a concentrated effort on the part of each of the filter manufacturers to put himself in a proprietary position, has led to considerable confusion in the specifying and using agencies. As a result of this changing design, a necessarily long lapse of time between order and use, and very poor marking of the shipped units, it is often almost impossible for the layman to tell what type a particular group of filters in the warehouse might be that are now ready for use in a particular area. With the very high cost of "hot changes" even in small (20 filters) systems, a wrong guess can surely be costly and again robs you of production time.

A further disturbing trend recently has been the high number of defective units received from the manufacturer. In prior years, 6 repairs out of 600 units installed was high and was usually due to accidental damage at installation. However, as recently as this past week almost 100 containers had to be opened and checked to get 20 usable units. Even then, half of the 20 had to be patched before they were installed. This is a lousy contingency to have to face on a tightly scheduled, hot maintenance job.

Therefore, gentlemen, once again I would like to exhort this group to put forth effort in all possible haste to reach a standardization in our specifications and to insist on any and all manufacturers meeting these specifications. It is essential that we restore the "final air filter field" to the position of integrity, dependability, and service it once held.

# Discussion

- Q. Is the life of glass filters appreciably less than asbestos type?
- A. Haven't had glass ones in long enough to tell you. We think they will be somewhat less.
- A. (L. Silverman Harvard) Think life would be about the same.
- Q. (G. J. Hurwitz Army Chem. Center) Would like to know whether you attribute the damage in these filters to storage or deterioration in storage or did you receive them damaged from the manufacturer?
- A. We are unable to say, we think we received them that way had a prior shipment last year with no shipping damage. March damage was 96 out of 600. Later had to open up 100 to get 20 usable ones. Boxes were undamaged think they left the plant that way.

- Q. Have you any idea how long you had them in storage?
- A. Some we have had as high as one year.
- Q. Is it conceivable that they were damaged in storage?
- A. It is possible, yes. In prior years we had no trouble, but were using a different type of filter material.
- Q. To what do you attribute the experience of those which were destined for Hanford and reached Rocky Flat? Why did they fail so quickly? How do you go about the patching process?
- A. Small holes were squirted with cement. When all else fails, you replace the unit later asbestos glass not showing the proneness to damage.

We have some very minute amounts of Hf coming through - glass filters are not so prone to hf damage, but also we do have a small amount of hf damage.

# A SUMMARY OF AIR CLEANING ACTIVITIES AT THE SAVANNAH RIVER PLANT

J. J. CROLEY, JR.

E. I. du Pont de Nemours & Company, Inc., Savannah River Laboratory, Augusta, Ga.

#### INTRODUCTION

Air cleaning activities at the Savannah River Plant involve extensive use of various types of air cleaning media ranging from liquid systems, such as caustic scrubbers, to sand filters. Like other AEC installations, the Savannah River Plant leans heavily on CWS-6 and all-glass fiber types of high efficiency filters.

#### DISCUSSION

Two groups of applications are considered in this discussion of air cleaning activities at the Savannah River Plant: (1) the cleaning of air for supply to indoor areas, and (2) the cleaning of exhaust air systems before discharge to the atmosphere.

#### 1. Air Supply Cleaning Systems

All air supplied to production and laboratory facilities is filtered through "Airmat" type filters. These filters are fire-proof. They consist of bonded glass fiber having an initial resistance not exceeding 0.10 inch water gage at a linear velocity of 35 feet per minute. Average dust arrestance shall not be less than 90% as determined by the Air Filter Institute Code Test Method. Filter frames are of heavy metal. It has been found necessary to change these filters on a frequency of only once per year in all installations. This media is exceptionally durable and has eliminated the problem of perforation under heavy load. Experience with this media at SRP has been very satisfactory to date.

Air supply to locations containing delicate instruments or sensitive laboratory procedures is often filtered through high efficiency filters of either the fireproof or fire-retardant type, depending upon the degree of fire hazard involved.

## 2. Air Exhaust Cleaning Systems

#### A. Reactor Installations

Air exhausted from reactor installations is not filtered, except to a minor degree for high level caves, but is discharged directly to the environment through a 250' stack. High level cave exhausts are filtered through high efficiency filters of the fire-retardant type. A constant monitoring program is utilized to measure the amount of radioactivity released to the atmosphere, particularly with regard to Argon which is produced when air is bled into the reactor systems during shutdowns, and tritium which is evolved from tritium-contaminated moderator.

## B. Separations Installations

Air exhausted from separations systems may be cleaned in several different ways before final release to the atmosphere. These methods are discussed as follows:

#### 1) Sandfilters

Sandfilters are utilized to clean air from several different canyon process streams. While efficiency data for this type of filter is somewhat controversial, Savannah River Plant experience has shown that such a filter can handle up to 300 curies of particulate contamination with an average decontamination factor of 99.95%. Pressure drop across the filter used for this example has increased only about 0.29 inch water gage or 6.52 to 6.81 inches water gage during a period of 4 to 5 years. Design specifications allow an increase of one inch water gage. The sandfilter is also used to back up other filtering systems utilizing fiberglas, caustic scrubbers, and dehumidifiers.

#### 2) High Efficiency Filter Systems

This type of air cleaning media is used extensively throughout separations installations in all locations where toxic pyrophoric metals are handled. Prefilters are used extensively with this type of filter to increase its life expectancy. A review of all high-efficiency filter installations was completed recently resulting in a change from fire-retardant filters to fireproof filters in fire hazard locations.

Contamination released to these filters may vary enormously, depending upon leaks and spills in such process areas as the sample aisles and hot gang valve corridors. Experience has shown that filtering efficiency for CWS-6 type filters varies according to service from about 95% for mixed fission products to 99.9+% for plutonium. The Health Physics Section maintains a close check on the efficiency of these filters by constant monitoring both before and after final filtration.

High efficiency type filters are also used to clean exhaust air from recovery facilities. Uranium dust is the primary contaminant. Collection efficiencies approaching 100% are now obtained in these locations.

## C. Metal Fabrication Facilities

Metal fabrication facilities employ local exhaust systems extensively for operations where uranium and its alloys are handled or processed. These local exhaust systems are then filtered through high-efficiency filter units of the CWS-6 fire-retardant or fiberglas fireproof type, depending upon the degree of fire hazard involved.

#### SUMMARY

In summary, current air cleaning practices at SRP utilize a variety of techniques directed at securing a maximum decontamination factor for radioactive gases and particulate matter. The most commonly used device is the high-efficiency package filter. All "critical" locations where a potential fire hazard exists employ fireproof type filters. Other filter installations utilize the fire-retardant, high-efficiency type filter as an economic factor, until the current inventory of these filters is exhausted. Future plans visualize the use of only the fireproof, high-efficiency filter at this Plant. Air cleaning experiences at the Savannah River Plant were good with no problems worthy of discussion.

# Discussion

- Q. (J. Murrow U of Cal.) You mentioned fireproof filters, just what are they compared to fire type?
- A. That is perhaps a term or nomenclature used specifically in SRA Divided filters into types fire type to stand 250° continuously and fireproof to stand 800°F. continuously.
- Q. Do you have any results on the silver plated tape tests at this time? Any other clues as to your activity bursts?
- A. Not yet. One of the largest ones we did have seemed to be about the time we had our largest fallout days and I would hesitate to even suggest that this was the cause. However, it would look a little bit in that direction.
- C. (W. King, PPCo) These questions interest me small increase about two times output similar to what you had. Have you removed the tape and analyzed it to see if it is fresh fission products or foreign fission pulled through the flush zone?
- A. Unless you sit right with it, you can't do it. Sometimes happens at night have tried to cut the tape and thought it was that very thing. Next time it was something else. We are completely at sea. The only reason we have noticed is that our old fuel oil background levels ran so high -- that weren't noticeable.

- Q. What is the type of filter just before the Venturi that leads to the stack?
- A. FG 100 after filters Dahlinger glass test type which should be getting better and better all the time because they've been in there about 9 years high temperature filter the after filters are doing a good job, we think.
- Q. (L. Gemmell, BNL) Is it a general practice to test all filters before installing?
- A. (H. Gilbert, AEC, Washington) Join the discussion on filters tonight and find out the answers.

# FILTER EFFICIENCY STUDIES AT GOODYEAR ATOMIC CORPORATION

P. DIAMOND

Goodyear Atomic Corporation, Portsmouth, Ohio

#### ABSTRACT

Studies were conducted at Goodyear Atomic Corporation to evaluate the efficiencies of air filters and to gather data which could be used as bases in establishing effective filter cleaning and oiling schedules. Two types of filters were tested under various conditions and corresponding efficiencies were determined. A direct relationship was found to exist between the degree of louver opening and filter efficiency. The resistance across filter banks was found to increase over relatively long periods of time.

This paper presents the methods employed during the studies and the results obtained which were used to determine filter efficiencies and establish effective cleaning and oiling schedules.

#### INTRODUCTION

Despite the fact that the viscous impingement filter was the original filtration device, a review of the literature reveals very little on the collection efficiency of this filter for atmospheric dust. Studies were conducted at Goodyear Atomic Corporation of two types of impingement filters in an effort to evaluate efficiencies under various conditions and to establish effective cleaning and oiling schedules based on both filtration efficiency and resistance measurements. The magnitude of the studies may be illustrated by the fact that a total of over thirty million cubic feet of air is filtered each minute in three process buildings at Goodyear Atomic Corporation. Figure 1 shows one of these process buildings and its air-intake louvers. There are sixty-two filter rooms throughout the three buildings, representing a total of 64,500 square feet of filtration area. The average air flow through the filters is 500 linear feet per minute.

Figure 2 is a diagram of air circulation in a typical process building where the filtered air supply is used to cool compressor motors. Some typical ambient temperature readings are also shown on the diagram.

### TEST PROCEDURES

Preweighed fiberglass filter papers were used in conjunction with Staplex hi-volume air samplers. Samples of the air were collected both prior to and following air passage

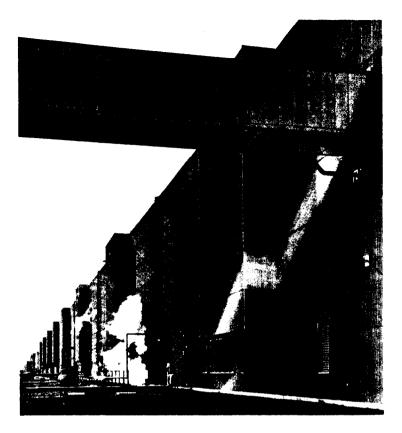


Fig. 1 — Typical process building showing air intake louvers.

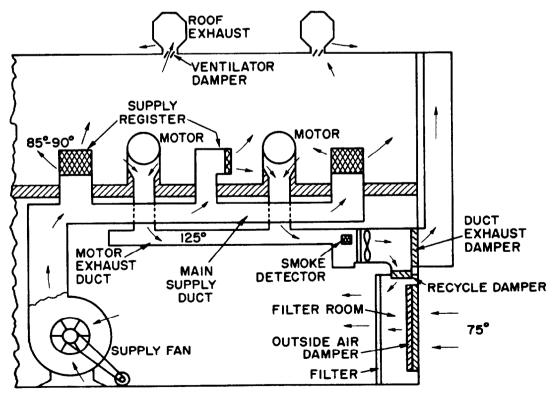


Fig. 2—Typical ventilating system.

through the filter banks. The filter papers were dried at 110°F. for one-half hour and weighed immediately upon removal from the laboratory oven. The drying process was necessary in order to stabilize moisture content. The papers were then placed in the hivolume samplers located on both intake and outlet sides of the air louvers and air was sampled at the rate of approximately 12 cu. ft. per minute. Following the collection of samples, the filter papers were again dried and reweighed. The increases in weights of both filters were used as bases for the determination of efficiency.

#### RESULTS

## FAR-AIR FILTERS

Results of the tests of Far-Air filters indicated an over-all average efficiency of 42%. Twenty-eight pairs of samples were collected; each sample being collected for about seven hours. As previously stated, the increase in the weights of the filter papers, before and after the filter banks, was used in determining the efficiency by weight of the filters.

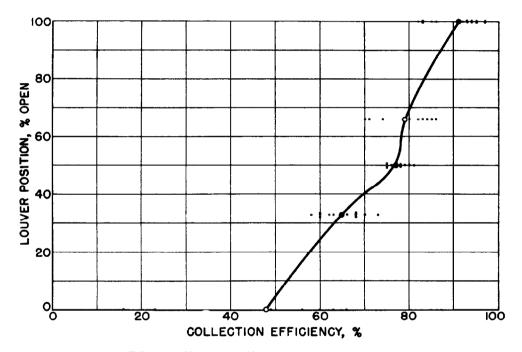


Fig. 3—Filter collection efficiency vs. louver position.

## **HV-2 FILTERS**

Fifty-four pairs of samples were collected to determine the efficiencies of this type filter. Louver openings were adjusted during each of the tests in order to permit the entry of a predetermined amount of unfiltered air. The over-all results of these tests show the following efficiencies:

Louver Position	No. of Tests	Average Efficiency
Closed	7	48%
1/3 Open	13	<b>65</b> %
1/2 Open	10	77%
2/3 Open	9	<b>79</b> %
Completely Open	15	91%

The individual test results are graphically presented in Figure 3. It appears that a direct relationship exists between the degree of louver opening and dust collection efficiency. This may be explained by the fact that as the degree of louver opening increases, more air, containing larger foreign particles, is admitted from outside the buildings. The filtering of these larger particles results in the greater efficiency.

#### CONCLUSIONS

Experience to date has indicated that during an eight-month period the average resistance across the filter banks increased from 0.08 to 0.20 inch of water. Based on the manufacturers' specifications to clean the filters at the 0.25-inch level, a cleaning schedule has been established on an annual basis. An oiling schedule has been established at three times per year. The most economical method for cleaning and oiling the filters at Goodyear Atomic Corporation has been found to be the direct removal of the filters and immersion in an alkaline dip tank. The filters are subsequently cold water rinsed, hot water rinsed, replaced, and finally re-oiled using SAE 40 lubricating oil. The estimated cost is 16 cents per filter.

Sufficient evidence has not been accumulated to evaluate fully the optimum time interval between filter cleanings, but the necessary background material has been gathered to permit better evaluation of data to be collected in the future.

# SUMMARY OF AIR CLEANING ACTIVITIES AT BROOKHAVEN

LEE GEMMELL

Health Physics Division, Brookhaven National Laboratory, Upton, N. Y.

#### ABSTRACT

A number of radioactive peaks have been seen on the dust monitor located in the cooling air of the BNL Research Reactor. These peaks are much more pronounced since the installation of the new enriched fuel in August 1958. Even though this radioactivity represents no health hazard to the environment, a study was made to determine the cause of these peaks. The results of this study were inconclusive and showed only that major changes in meteorological conditions bring changes in peaks but no specific correlations have been made.

A study is in progress to test the chemical combination of iodine and bromine on silverplated copper gauze at different temperatures. No results are available.

The Meteorology and Health Physics Divisions are studying the size, shape, concentration and height of the cooling air plume from the research reactor under various weather conditions using the radioargon in the air plume as the trace element. Preliminary results look satisfactory.

\* \* \* \* \*

The largest and perhaps the most interesting and challenging air cleaning problem at Brookhaven is in connection with cleaning the cooling air of the Research Reactor. As you know, this reactor is air cooled, graphite moderated and, since last August, the original natural uranium has been replaced by an enriched fuel. To cool it requires approximately one million pounds of air an hour. This air is pulled with a pressure drop of about 50 inches of water through inlet filters, the reactor, and exit filters by five 1500-horsepower fans and exhausted from a stack 350 feet above the surrounding terrain.

The inlet air filters are deep pocket fiber glass (American Air Filter RG-100) and the exit air filters are woven glass fiber filters (Dollinger Glass Tex). The inlet filters are reasonably satisfactory for removing atmospheric dust. However, many particles in the sub-micron size pass through in the air stream. The Health Physics Division has installed a continuous dust collector for sampling the cooling air at the base of the stack. Figure 1 shows the cooling air system of the Brookhaven Reactor and Figure 2 shows instruments used in monitoring air for particulate activity.

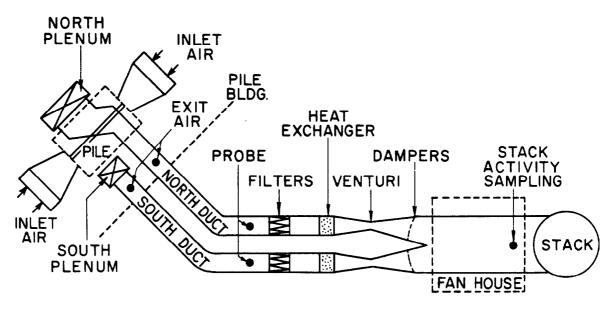


Fig. 1—Cooling air system, Brookhaven Reactor.

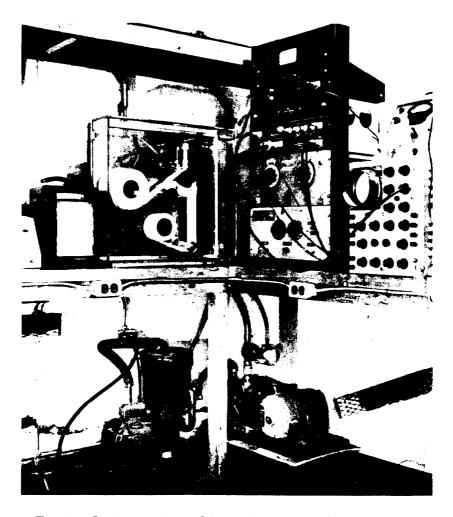


Fig. 2—Instruments used in monitoring air for particulate activity.

Of course, the principal radioactive contaminant in the cooling air stream is Argon-41. Even though there is very little Argon in air, approximately 15,000 curies of A<sup>41</sup> are discharged up the stack each day. The height of the stack, the relatively short half-life of the radioargon (110 minutes) and the tremendous dilution by the atmosphere, causes no problem in disposing of it safely in this manner. The amount of A<sup>41</sup> produced depends on reactor power level, amount of air moved and the length of time the air remains in the flux so tends to be fairly constant and predictable. The particulate problem, however, is more interesting because of certain natural and man made disturbances causing dust that affects the over-all radioactivity of the cooling air effluent.

Prior to the replacement of the natural uranium fuel last August, there was always a certain small amount of mixed fission product contamination from small fuel element leaks into the air stream. The dust monitor in the stack always detected a small "background" of this activity and tended to remain fairly constant. This amount of radioactive dust represented no hazard as far as the local environment was concerned.

After the change over to the new fuel, the same dust monitor displayed some rather sharp peaks that lasted for periods ranging from an hour or two to as much as three days. It was thought that these peaks were likely caused by possible uranium contamination on the outside of the new fuel, possibly by small blisters reaching through the cladding into the U235 alloy, or the scrubbing of fission product debris left over from the old fuel. The enriched fuel from one of the central holes was removed for routine replacement and it was then discovered that one of the elements was badly damaged. Following removal of the damaged element, there appeared to be less frequent peaks. This improvement may have been due to the removal of the damaged element or merely a coincidence.

Radioactive peaks still occurred although less frequently. A dust collector and monitor was installed in each exit air duct before the exit filters to see if the activity was coming from only one side of the Pile. Both monitors showed the same rises at the same time, indicating that something must be getting into the air stream before entering the Pile. It was at this point that automobile exhausts were linked to the rises in activities. If a truck or passenger car were left running in the general area of the inlet air intake, the activated exhaust would show on the dust monitor. This discovery looked promising as the answer. However, within a week, another peak appeared with no man made disturbance as far as could be ascertained.

A short time later, one of the Operations personnel noticed that a peak was coincidental with a shift of the wind to the south. The wind direction changed abruptly from the northwest about 15 minutes before the trace appeared. From windspeed data, the sea air from the south seemed to be perfectly timed for the peak. However, on later occasions, more peaks occurred without sea breezes.

Later, on a very humid foggy day, a large sharp peak appeared with nothing mentioned before as a possible contributing factor. The theory advanced this time suggested that moisture penetrating the inlet air filters was carrying dirt from the inside surface of the filter into the air stream. The next time this same atmospheric condition appeared, there was no peak. Since then, several cases have been examined and each time there seems to be less and less correlation.

If atmospheric polution is causing these abnormalities, then a sample of the atmosphere should be taken at the same time the peaks occur in the cooling air and analyzed spectrographically. This procedure was

set up by providing an automatic triggering device that would turn on another air sampler for sampling the atmosphere when a peak showed in the Pile stack. These samples have shown nothing unusual to date. The dust monitor peaks are showing less frequently during the late spring and summer so it will probably be necessary to wait until fall for more data. The cause of these radioactive peaks at the moment is unknown. Major changes in meteorological conditions bring changes in peaks, but no specific correlations have been made.

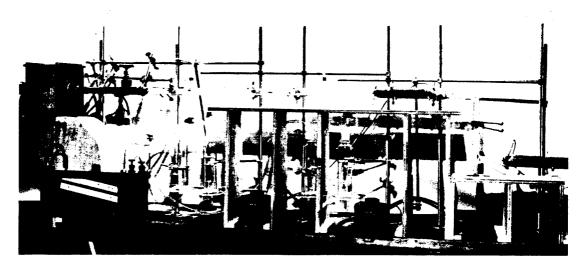


Fig. 3—Apparatus for the laboratory experiment.

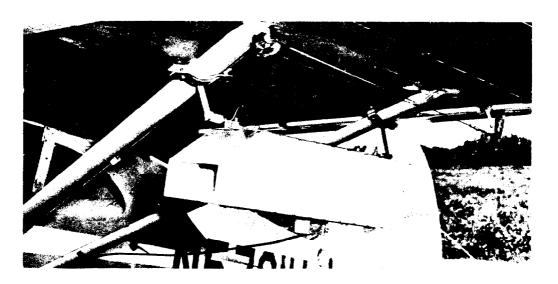


Fig. 4—Geiger counters mounted in aluminum box attached to underside of plane wing.

It is obvious that contaminants in the air, even though in very small concentrations, can be detected upon passing through the neutron flux in the reactor. The additional sensitivity of the reloaded Pile is due to the fact that since the new fuel reloading there is an area of considerably higher specific power, the flux is up by a factor of 3 or 4 and the air stays in the Pile longer.

The staff at the Harvard Air Cleaning Laboratory has been interested in testing the chemical combination of iodine and bromine on silver-plated copper gauze at different temperatures. Even though there are no routine problems or hazards connected with radioactive iodine or bromine at Brookhaven, our Reactor Operations Division is also interested in the project as an aid in cleaning up the cooling air effluent in case of a reactor accident. The test calls for the irradiation of a small piece of fuel plate, melt it quickly in an air stream and measure the amount of radioactive iodine and bromine on the silver-plated copper mesh in the stream. Figure 3 shows apparatus for the laboratory experiment.

The Meteorology and Health Physics Divisions are studying the size, shape, concentration and height of the cooling air plume from the research reactor under various weather conditions. To do this, an airplane flies a course back and forth through the cloud at varying distances and heights from the reactor stack. Geiger counters are used as detectors and are mounted in an aluminum box attached to the underside of the plane wing. (Fig. 4) The geiger tubes are sensitive to  $\beta$  particles and relatively insensitive to  $\gamma$  -rays. The air flows through the box at approximately the speed of the plane and the GM counters see primarily the activity in the box. The geiger pulses are recorded on a tape recorder and will be analyzed later on a multichannel type machine. It is hoped to get within 50 feet of the actual plume size. Preliminary results look satisfactory.

The Meteorology Group is also cooperating with the Health Physics Area Survey Group in an analysis of the effect of the use of the enriched reactor fuel on radiation in the local vicinity. The analytical procedure involves a careful separation of those hours in which a given monitoring station was unaffected by the reactor plume and those in which such an effect was certain or possible. This, coupled with appropriate radiological data from the Health Physics background monitoring stations and reactor operations data, should provide rather substantial detail on the problem after a study of about 6 months of data.

# AIR CLEANING PRACTICE AT U.K.A.E.A., ALDERMASTON

F. J. HALL and S. E. SMITH United Kingdom Atomic Energy Authority, Aldermaston, Berks, England

## I. Air Cleaning Requirements

Air cleaning facilities are installed at Aldermaston in the ventilation systems of laboratories and workshops where radioactive and toxic hazards arise, in particular from experiments involving the handling of natural and enriched uranium plutonium and beryllium.

The Health Physics Branch has a dual responsibility, to limit the exposure of staff who work on toxic or radioactive materials to a safe level, and to limit, and record, the quantity of activity discharged to atmosphere from the site as a whole. In pursuance of these ends, they specify the standard of room ventilation and cleaning of extract air which is required for each particular operation. They also operate monitoring services to check that these ends are achieved.

Room ventilation requirements range from 5-20 air changes per hour for active and toxic laboratories, to 10 air changes per hour for semi-active areas such as change-rooms and stores. Air is usually supplied to these areas via a filtered plenum system which promotes cleanliness within the building and relieves the dust load on the extract filters.

Handling of exposed plutonium in all forms and of finely divided beryllium and uranium, is carried out in sealed glove boxes under a negative pressure with a recirculating inert atmosphere. Provision must be made for filtration of the High Pressure Extract discharge to stack which comes into operation in the event of a pressure rise in the boxes. High efficiency filters are installed on this H.P.E. extract, and also on the inert gas exit lines to confine contamination to the glove box installation.

Processing of beryllium and enriched uranium in massive forms is carried out in air hoods or fume cupboards which are extracted at such a rate as to provide a flow of at least 200 LFM through all openings. These are often equipped with a local low efficiency separator (coarse filter, cyclone or scrubber, according to circumstances) which facilitates the recovery of valuable materials and lightens the load in the main filter. For the average laboratory or workshop, the total extract from the installed machine boxes or fume cupboards is sufficient to supply the required number of room air changes per hour.

The combined extract air is required to be filtered before discharge to the stack, the policy being that contamination is reduced to a safe level by the cleaning system, plus the dilution achieved before reaching ground level by discharge from a stack of a certain height.

### II. Design of Air Cleaning Systems

The specification and design of filtered extract systems is the responsibility of the Chemical Engineering Branch, whose work in this field falls into two main categories:

- (a) design of air cleaning systems for spcified applications in new or existing buildings, and
- (b) development and testing of new types of filters and other air cleaning devices.

#### 1. Filters

The earliest installations at Aldermaston employed commercially available cylindrical filter units, about 5 ft. long x  $8\frac{1}{2}$  ins. dia., in which the filter medium was wrapped twice round a central perforated tube; the canister is welded up after packing (and disposed of with the filter after use). The medium employed is resin-coated wool, developed for removal of particulate smokes in respirators; these filters have a throughput of 180 cfm at a pressure drop of 0.5 - 0.8 ins. w.g., and a methylene blue penetration of 0.05%.

It was known that the filtration efficiency of resin-wool filters decreases during prolonged continuous operation, and to some extent during storage. Where continuous high efficiency was required, the resin-wool was replaced by a double-wound pad of asbestos-wool (mixture containing about 18% asbestos,) which gives a methylene blue penetration of 0.001%, but with a throughput of only 36 cfm at a pressure drop of 1.0 - 1.2 ins. w.g. In some instances an electrostatic precipitator was included.

This system, which is efficient but expensive in space and installation costs, has been replaced in later buildings by a pleated pad type filter, developed by the Chemical Defense Experimental Establishment, Porton, and supplied to U.K.A.E.A. specifications, employing a 50:50 cotton-asbestos filter medium, used either along or in conjunction with a pleated glass-wool prefilter made up from one layer of fine superimposed on one layer of superfine glass-wool, which extends the life between changes of the main filter. Throughput of these cotton-asbestos filters is 200 cfm for an 18" cube filter unit, at an initial pressure drop of 1.1 - 1.5 ins. w.g. and a methylene blue penetration of < 0.003%. These filters will withstand a temperature of 120°C before the cotton chars although at this temperature the asbestos would probably still remain as an effective filter.

A technique has been developed for replacing the prefilter medium in situ under highly active working conditions, (i.e., by working in "frog" suits); when the main filter is clogged, the whole canister is disposed of.

For the lower efficiencies required for work with uranium, a similar unit was developed, with one can 18" x 18" x 18" in which the main filter medium was a double layer of AA Grade with a prefilter of one layer of Fine Grade Fibreglass with pressure drop of 1.5 ins. w.g. at 330 cfm, and a methylene blue efficiency of 0.35%.

More recently pleated paper type filters have become available, which are still more compact. Originally the commercial units of this type employed asbestos paper, containing 5% of asbestos fibre, which is highly susceptible to the effect of moisture in the filtered gas and has only a very limited degree of fire resistance. Fireproofing of cotton-asbestos and asbestos paper has been tried with some success, but it is not considered by us to be a sufficient protection.

We are considering the adoption of pleated glass-paper units which are highly resistant to moisture and still effective after completely wetting and drying out again, and, with choice of suitable containers and cements, can be operated continuously at temperatures up to 500°C. A 24" x 24" x 12" unit has a throughput of 1000 cfm at 0.5 ins. w.g. pressure drop, but with a methylene blue penetration on only 0.01 - 0.03%. Two units can, however, be placed in series in a 24" cube

canister to give penetration of  $\langle 0.003\%$ , and it is considered at present that this type of installation represents the most satisfactory solution to most filtration problems. It can also, at the expense of a slight loss in high temperature resistance, be made as an acid resistant unit.

The actual filter unit is pleated and sealed into a disposable metal frame, with separators between the folds made of pleated aluminum foil and, in the acid resistant one, resin impregnated fibreglass. This has been done by the firms concerned to our requirement. The units are pressed against the sealing faces of the canister by a wedge action.

### 2. Design of Installation

All main filters and fans are housed in a separate ventilated filter room within the building served, which, where high activity is involved, is a "purple" area, i.e., in which any operation such as filter changing must be carried out in "frog" suits with air line supply. Extract ducts are at present designed to give flow rates of about 1800 L.F.M. which has proved satisfactory in limiting deposition of particles within them, but we are not certain that this is the optimum in all cases, and should be interested to hear of U. S. practice.

## 3. Local air treatment prior to filtration

In many cases, the extract system from boxes and from hoods incorporates a local treatment which provides additional decontamination of highly active extract air, permits recovery of valuable materials, decreases the load on the main filters and lessens the danger of settling of larger particles in ducts. It may also act as a spark arrestor in certain cases, to decrease the danger of fire in the extract system.

Such installations vary according to the size of the airborne particles, and with the work being carried out in the enclosure. They may be simple, inexpensive and easily replaced glass-wool mats, high efficiency cyclones (wet or dry), bag filters, wet scrubbers or acid scrubbers.

## 4. Plenum Air Treatment

Most active buildings are provided with a plenum supply for temperature control. This is normally filtered for the reasons stated earlier, the local atmosphere dust loading is about 0.07 mgm/cu.m.).

Early installations were electrostatic precipitators followed by Vokes "Kompak" cotton fabric filters, a filter unit 12" x 24" x 18" giving a pressure drop of 0.2 ins. w.g. at 600 cfm. Efficiency on normal atmospheric dusts was about 90% in the precipitron and about 30% in the Kompak, giving an overall efficiency of 93%. Electrostatic precipitators however, are subject to occasional bursts with 100% penetration, or even to discharges in which the air loading is increased above input.

A later development was therefore the Kompak Composite filter in which a Kompak cotton fabric filter was preceded by a glass fibre prefilter. This gave a pressure drop of 0.5 ins. w.g. at 400 cfm with a methylene blue penetration of 2%.

Currently recommended systems consist of Vokes '44' pleated asbestos paper, 24" x 24" x 12" units preceded by glass fibre units which act as roughing filters and spark arrestors. This system has a pressure drop of 0.75 ins. w.g. at 1000 cfm and methylene blue penetration of  $\langle$  2.5%.

In all systems, the air is preheated to bring it below saturation with respect to moisture before passing to the filters.

#### III. The Work of the Filter Development Section

## (a) Specification of filter systems for new or modified buildings

The building design engineer is supplied with a detailed chemical engineering specification with flowsheets, which satisfy all the requirements of User, Health Physics. Safety etc.

## (b) Filter Testing

All filters are tested individually before installation in any active system in a test rig; the Filter Development Section uses one for experimental work. Pressure drop and filtration efficiency are determined at the designed flow rate. Filtration efficiency is normally tested by the methylene blue penetration test which is laid down in a British Standard Specification.

Where special filters are needed for local extract points, additional tests may be carried out with heavier loadings of standard test dusts - usually alumina or talc.

## (c) Development of New Filters

This has gone on over many years and we think that at present the best available answer to our problems is the pleated glass paper filter, as a high capacity, fire-proof heat-resisting and moisture and acid-proof filter for all active and toxic extracts; where high efficiency filtration is necessary, two are used in series. The asbestos paper unit with glass fibre prefilter is accepted for plenum filtration as it is cheaper.

## (d) Assessment of Filtration Costs

This has been carried out in detail for certain buildings in use, and provides data for assessment of costs of filtration in new buildings and provision of estimates for ventilation system.

## (e) Investigation into airborne contamination

Determination of dust loading and particle size distribution under working conditions to determine filter loading rates and determination of efficiencies required.

#### (f) Review and replacement of older installations

Some systems installed during the last ten years are under review at present, and consideration is being given to the advantages in economics, efficiency and safety which can be achieved, together with release of building space, by replacing them with more modern types of filters. The work involved in making such changes under contaminated conditions is being looked into.

## Discussion

- Q. (G. J. Hurwitz, Army Chemical Center) In view of the fact that use of p.o.p.is pretty well standardized in this country for more than 10 years, I'm surprised you are still using methylene blue in Britain. If it is going to be used any length of time --?
- A. We find methylene blue easier to handle and results more produceable \_\_\_
- C. We understand and that's why we are interested in this. It doesn't take so long to get answers.

# SUMMARY OF IMPORTANT AIR CLEANING ACTIVITIES AT FMPC, FERNALD, OHIO

C. H. STEVENS
National Lead Company of Ohio, Fernald, Ohio

Our criteria for industrial ventilation are similar to most other AEC installations; namely, (1) protection of the health of the worker by providing environment well below the M.A.C., (2) reduction of uranium losses through exhaust stacks to an absolute minimum and (3) keeping maintenance and downtime on ventilation equipment at the lowest possible levels.

This talk will briefly discuss some installations we have recently completed which I believe will be of interest to most of you. One job involved collection of very fine particles which caused us considerable difficulty in maintaining constant pressure drop across the dust collector bag. Another project involved the installation and operation of a prototype drumming station which has virtually eliminated dust leakage to the room while filling drums. A third installation dealt with a method of collecting dust from a booth where uranium ingots are separated from their molds. The last job I will mention will deal with the burning off of up to 5% enriched sludges in our Pilot Plant. We will also mention a few of the ventilation projects we are presently working on and expect to install within the next year or so.

In our Green Salt Plant we reduce UO3 to UO2 by passing hydrogen through UO3 in a reactor. (Show Slide). The excess hydrogen used is burned, and the off-gas is passed through a bag collector where any fine material picked up by the hydrogen stream in its passage through the reactor is collected. We recently converted from screw reactors to fluid bed reactors, which resulted in higher loadings to the dust collector as well as finer dust particles. Our measurements indicated a loading of 2/10 of a grain per cubic foot of air. About 70% of the dust particles are finer than 5 microns and about 30% finer than 1 micron. Although this grain loading is by no means considered heavy in normal dust collection operations and we were only operating at an air to cloth ratio of about 12 to 1, we obtained pressure drops across the collector bags

of 6 to 8 inches, with that pressure drop increasing by the hour. Periodic shutdown of the collector and handshaking of the bags was necessary to keep operating.

We have succeeded in remedying the situation by doing the following:

Utilizing a small collector located nearby in parallel with our H<sub>2</sub> off-gas collector and thus dropping the air to cloth ratio to about 8 to 1. We also revised our hooding as much as feasible to reduce air volume requirement and still get adequate collection. We further installed truncated cones on top of the bags to prevent recirculation of the fines. In addition, we went over these collectors with a "bine tooth comb," checking proper blow ring center distances, carriage travel speeds, and other mechanical facets of collector operation. Pressure drop is now uniform somewhere around 4 to 5 inches across collector bags.

Another project was concerned with obtaining proper dust control at points where we are filling 55-gallon drums. We, therefore, decided to design and install a prototype drumming station which, if it works satisfactorily, will be used throughout this project. This slide shows this prototype drumming station just installed. As you can see, it is basically an enclosure with adequate windows and floodlights; we have also provided a semicircular ventilated hood around the drum. The station itself is also ventilated. We have allowed enough space in the drumming station so that the operator can move the drum away from the feed spout and lid the drum inside the enclosure. An air dust survey just completed shows excellent control at this drumming station; the counts taken at this station were practically the same as the general background level.

Another project just completed and now operating is a new ingot separation booth in our Metals Plant, where the ingot is separated from the mold. This booth is ventilated and the exhaust ducts are brought into an existing Mikro-pulverizer dust collector. There are existing ducts, brought in to the top of the collector by a specially sized manifold, as shown on the slide. To modify this manifold to provide for the additional air quantities required for the ingot separation booth ventilation would demand expensive duct changes. We, therefore, brought a new duct into the hoppers of this collector, as shown on this slide. Thus, in effect, most of the air enters the top of the collector, with about 25% of the air entering the bottom. No difficulties have been experienced as a result of this somewhat unusual method of air entry. The collector operates at an air to cloth ratio of approximately 13 to 1 and a bag pressure drop of 3 to 4 inches.

In our Pilot Plant, we have had the problem of burning off sludges of up to 5% enrichment. The fumes from this

burning process are sticky and oily and generally difficult to collect. We tried some Cambridge "absolute" type filters preceded by a fiberglass prefilter, but found that the "absolute" filters plugged up rather rapidly. We have since replaced these filters with two banks of American Air Filter Company's PL-24 units of fiberglass media. Due to their larger dust holding ability, they have operated successfully for some time with no noticeable stack losses.

We are now working on a number of ventilation projects, such as a new off-gas system for the rotary kiln. As shown on this slide, the present system is a relatively complicated one and has a lot of ducts and equipment that not only could plug up but do plug up. We, therefore, propose a much simpler exhaust system, as shown on this slide, which basically involves only a few feet of ductwork with adequate cleanout doors, and a caustic scrubber with hopper bottom. Another project involves ventilation from our digestion tanks used to dissolve uranium scrap in the Scrap Plant. This ventilation is essential to vent hydrogen fumes generated by the digestion process. Unfortunately, the exhaust air also draws off hydrochloric acid fumes which we are planning to collect in a Chemical Construction Company Pease-Anthony Venturi Scrubber. This scrubber was picked for its inherent simplicity and consequent ease of operation and maintenance. Other installations being designed now involve additional ventilation for our green salt canning stations using a Wheelabrator Corporation Dust Collector, an off-gas scrubber for a sludge furnace in which grinder sludges are burned, ventilation for a Williams Crusher and allied equipment, general purpose bag collector for our Pilot Plant operations in which up to 5% enriched dusts will be collected, and a new continuous-automatic bag collector for our low enrichment metal remelt operations.

# STACK SAMPLING PROCEDURE AT THE NATIONAL LEAD COMPANY OF OHIO

ROBERT H. STARKEY

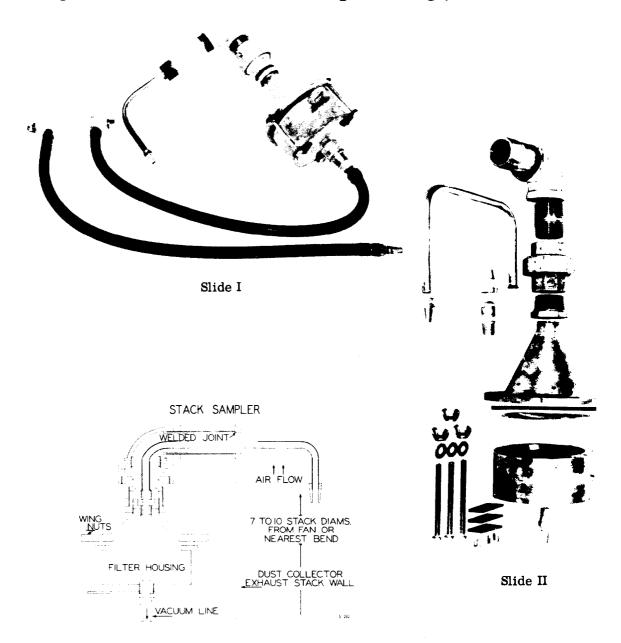
Health and Safety Division, National Lead Company of Ohio, Cincinnati, Ohio

As at all AEC installations, all of us here are interested in the amounts of material being exhausted to the atmosphere. Ventilation equipment and dust collecting equipment represent an important investment at all AEC sites and we, as health and engineering people, are quite interested in seeing that this equipment is operating as efficiently as possible.

We decided to make a study to determine the efficiency of the dust collectors at the Feed Materials Production Center. As a result of this study, we developed a stack sampler and sampling procedure for the purpose of monitoring as many of our stacks as possible. The stack sampler is shown in Slide I, and I will describe the procedure we developed. It might possibly have a use at your site.

Presently we have approximately fifty of these samplers operating on a continuous basis. These cover all dry stacks. We have found that the results obtained from this program have been a very important tool in increasing the efficiency of the operation of the dust collecting systems in our plants. A preventative maintenance program on dust collectors was just getting under way at the time our stack sampling program was instituted. The stack losses which we reported, along with their accompanying monetary worth, were a valuable aid in speeding up the preventative maintenance program. The over-all dust collection inspection and maintenance program has improved to the point where we are now losing only 15% of the material that was being lost at the time of the original installation of the stack samplers. You can see, therefore, that the stack sampling program, in addition to greatly reducing the quantity of radioactive material released to the environs, has more than paid for itself in the intrinsic value of the material collected. Another indirect by-product has been the improvement of the ventilation provided on equipment as a result of the more efficient maintenance and operation of the dust collection equipment.

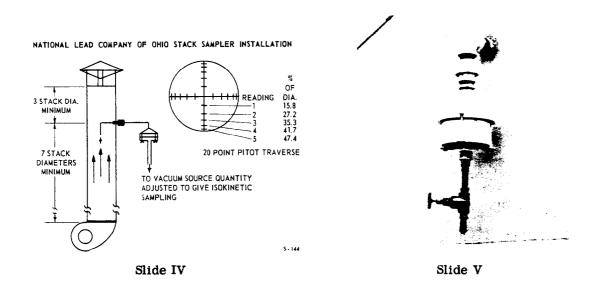
I would like to explain briefly the stack sampler developed, the procedure followed, and the use made of the results obtained. Slide I shows a picture of the stack sampler of the type presently being used. A number of changes have been made since the original design, all of



Slide III

which make the installation and sample changing easier. Notice the rotameter attached. I will explain more about its use later. Slide II is a disassembled view of the sampler and all of its integral parts. Slide III is a detailed sketch of the sampler showing the critical dimensions.

Slide IV is a diagram of an ideal stack sampler installation, and Slide V is a picture of an actual installation. This is typical of the other installations with the one exception that it is one of the few installed on the roof of a building. Most of our samplers are installed inside the buildings, inasmuch as the type of roofs make sampling thereon impossible. Slide VI describes the procedure for installing the sampler in a stack.



## SLIDE VI

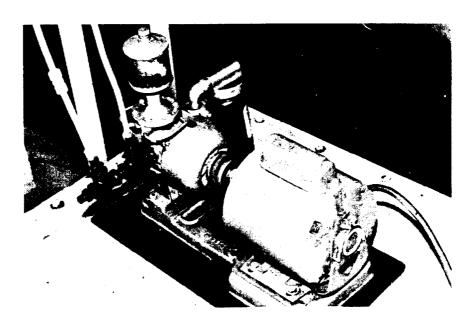
Procedure for Installation of Sampler in Stack

- Drill hole in stack best possible location inside building.
- 2. Traverse stack determine average and centerline velocities. (This should be done bi-annually so appropriate corrections can be made.)
- 3. Have sampler constructed for centerline sampling.
- 4. Determine best source of vacuum, either Hi-Vac unit (Slide VII) or Gast Pump (Slide VIII).
- 5. Install sampler probe in center of duct and parallel to line of air flow.
- 6. Determine volume of air needed for isokinetic sample.
- 7. Adjust flow through gate valve with rotameter attachment.
- 8. Remove rotameter.

Slide IX shows the procedure followed for changing the sample. The samplers are visually checked a minimum of once each month and changed if sufficient material for weighing on a beam or platform balance (0.1 gram) is found on the pleated filter. If insufficient material is collected, the filter is allowed to remain in the stack until such time as enough material has been collected for analysis. When a sampler is not changed, the loss for the particular stack is recorded as non-detectable for the particular sampling period. In addition, samples are changed following the release of unusual amounts of material from the stack due



Slide VII



Slide VIII

to damaged dust collector bags or a malfunction of the collector. The production personnel notify us immediately following an incident and we install a new filter.

## SLIDE IX

## Procedure for Changing Sample

- l. Disconnect vacuum line from sampler.
- 2. Insert rotameter and check flow.
- 3. Loosen wing nuts and disassemble sampler.
- 4. Remove filter and place in pre-numbered plastic bag.
- 5. Insert new pre-numbered and pre-weighed type "S" filter.
- 6. Assemble sampler and tighten wing nuts.
- 7. Again check sampling rate and make necessary adjustments.
- 8. Remove rotameter and connect vacuum source to sampler.
- 9. Submit sample for analysis.

In the laboratory the following procedure is used:

## Analytical Procedure

- 1. Pre-weigh and pre-number pleated filter and respective plastic bag prior to use.
- 2. Again weigh following use to determine gross weight of material collected.
- 3. Run wet chemical analysis of material collected to determine per cent of uranium.
- 4. Report both gross weight and per cent of uranium.

Slide X shows the calculation necessary for determining the pounds of uranium lost from each stack over any sampling period.

#### SLIDE X

$$\frac{V(ave)}{V(C_L)}$$
 x  $\frac{R(s)^2}{R(p)^2}$  x  $\frac{1}{454}$  x Ug = U loss - lbs.

V(ave) - Average stack velocity (fpm)

V(CL) - Stack centerline velocity (fpm) R(s) - Radius of stack - inches

R(p) - Radius of probe - inches
Ug - Uranium collected in grams

In summary, I have described the method that we use for estimating the material being exhausted from the stacks at our plant. It is fully realized that this is not an exact method in any sense of the word; however, we have conducted some preliminary tests which indicate the procedure is accurate within plus or minus 20%. We have found it a very worthwhile tool in checking the operation of our dust collectors and feel that possibly it has a use throughout industry. It can only be used where relatively large amounts of material (in the order of a pound) are being exhausted over a reasonable period of time; thus, it is probably of no use where absolute filters are used or on other types of systems exhausting only very small quantities of material. However, I do feel that where appreciable amounts of material -- whether radioactive or non-radioactive -are exhausted, this method can be profitably used. Actually an adaptation of this stack sampler can very well be used for determining the loading on dust collectors by installing it on the inlet side of the collector. By using both the inlet and outlet sampler simultaneously, an efficiency study can also be carried out.

If any of you are interested, I would be only too glad to furnish the necessary drawings and equipment specifications for this sampler upon request.

# AIR CLEANING AT THE KNOLLS ATOMIC POWER LABORATORY

T. T. POREMBSKI

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## Abstract

The construction of a new \$30,000 facility for the sole purpose of safeguarding against the spread of radioactive iodine has been started at the Knolls Atomic Power Laboratory. This facility is in addition to the many air cleaning facilities at KAPL which have been reported at previous AEC Air Cleaning Conferences.

Well over 475,000 cfm of laboratory air is cleaned at KAPL prior to discharging the air to the environs. The air is first introduced as 100% outside air thru air systems equipped to filter the air with self cleaning of throwaway filters. It is again filtered before it is discharged to the atmosphere. Over 350 CWB filters are employed in the exhaust systems. Multiclone and deep pack filters are also used.

A need for the control of radioactive iodine vapor in the Radioactive Materials Laboratory has created a new problem which has become unique to our operations and calls for a filter media which will retain gaseous matter as differentiated from particulate matter.

This report deals with the description of the newly designed activated carbon and stack system which will handle the radioactive iodine vapor and will be operated in conjunction with the existing systems in the Radioactive Materials Laboratory.

The new system will consist of a 17200 cfm fan and filtering facility and will employ 24 - 24" x 24" commercial activated carbon filters operating together with a 105 foot stack. The overall radioactive iodine concentration reduction factor will be in the order of  $4 \times 10^4$ .

In the event of a radiation incident or equipment failure back up features are included to safeguard against the spread of radioactive contamination.

Introduction - At KAPL there are 6 laboratory functions which call for the use of environmental air cleaning. The areas are shown on Sketch Y-A-1757 and are designated as follows:

- Special Nuclear Materials Machine Shop Bldg. D-3, D-4. (Multiclome & deep pack filters)
- 2. Chemistry Physics and Metallurgy Labs Bldg. E-1, G-1, G-2, A-3. (CWS filters)
- 3. Radioactive Materials Laboratory Bldg. E-2, E-3, E-4. (CWS filters)
- 4. Nuclear Reactor Components
  Inspection & Quality Control
  Bldg. Q-4, Q-5. (CWS filters)
- 5. Waste Disposal Bldg. H. (CWS filters)
- 6. Zero Power Reactors
  Bldg. F. E-5. (CWS filters)

Because of the close proximity of the buildings and the location of KAPL in a suburban area positive control of effluent gases is essential.

The Radioactive Materials Laboratory has presented a radioactive iodine problem which has made it necessary for the design of the iodine vapor system. Specific reference will therefore be made to the control of radioactive iodine.

The new facility will allow immediate sectioning and chamical-and corrosion testing of irradiated samples which in the mast could not be handled until the samples had sufficiently decayed and thus rendered safe to handle within the available facilities.

As stated above the new facilities will be added to the existing ventilation systems now serving the Radioactive Materials Laboratory. The purpose of the facility will be to fulfill the following needs, applicable to any radioactive materials processing laboratory.

- 1. Protect the laboratory personnel from any contact with contaminated materials whether such materials are in the particulate or vapor form.
- 2. Assure sufficient stack gas contaminant reduction to render the effluent gases harmless to the environs.
- 3. Provide safeguards which will assure continuity of operation and effective protection against the spread of contamination in the event of an incident.

In order to meet these needs the following critera are essential:

- 1. Introduce a tempered air supply into the building which will replace the air exhausted at the work areas.
- 2. Maintain uniform draft free air flow within the building in the direction of the contaminated areas. The building should be under a slightly negative pressure. The negative pressure

should become progressively greater in the direction of the hot work area until the highest negative pressure is attained where the hot work is performed.

- 3. Clean the air before it is discharged to the atmosphere.
- 4. Provide duplicate equipment if this is necessary to assure reliable operation. This is particularly applicable to fan equipment which is subject to breakdown.

The existing equipment at KAPL does not fully meet the requirements of items 2 and 3. Radioactive iodine might be discharged unless delays are imposed on the processing of radio iodine samples to allow them to cool off. There is also a chance of losing the continuity of air flow in the direction of the work within the cells in case of an incident. This latter inadequancy will be explained more fully after the existing facilities are described.

### Description of Existing Facilities

The RML building measures approximately 72' x 140' x 24' high. Here several hot laboratory functions are performed all of which are primarily associated with the examination of irradiated samples.

Air is supplied to the building with two central plant air units on a once thru basis. Both units are equipped with 2" thick ribre glass filters and together the units have a capacity of approximately 12000 cfm. Air is discharged from duct openings high up under the roof in order to avoid air turbulence at the occupancy levels.

Air is exhausted from the work areas with several individual exhaust systems each of which is equipped with a fibre glass prefilter followed by a CWS filter. From one (1) to six (6) such filters are installed ahead of each of the exhaust fans. The filters and fans are located on a 12' high balcony which runs along the length of the building. Discharge stacks rise straight up thru the roof from the fans to an elevation of 50 feet above ground level, approximately 25 feet above the roof.

#### A summary of the air flows is given:

 Autoclaves (in adjoining Cask Storage Bldg., 400 cfm fan in RML Bldg.)

2.	Localized Building Exhaust,	6,000
3.	Chemistry Hoods,	1,000
4.	Storage Vault,	200
5.	Decontamination Chamber & Room (in adjoining Bldg.)	2,800
6.	Cells 1 & 2,	3,000
7•	Isolation Boxes within Cells 1 & 2,	500
8.	Cells 3 & 4,	2,000
9.	Isolation Boxes within Cells 3 & 4,	300
10.	Ceil 5 & 7,	600

11. Cell 6,

300

12. Cell 8 (in adjoining Bldg.)

100

12 Stacks - Total Air Flow, cfm

17,200

Iodine work is primarily done in cells 1 thru 4, however it may be carried on to a degree in any of the other ventilated areas.

Since the optimum safety is required in cells 1 thru 4 the manner in which these cells are ventilated will be outlined in detail.

Figure No. 1 shows a diagrammatic view of the way in which the hazardous gases and dusts are contained within the work area. The work is performed within a isolation box consisting of an enclosure made of transparent polyethylene plastic sheets taped together nearly air tight. The boxes are about 3' x 3' x 3' in size and up to 3 or 4 isolation boxes are housed within a cell.

Proper operation of the ventilation depends on such factors as:

- Assurance all isolation boxes are sealed up. Short circuiting of air due to an open box or branch duct may interrupt the adequate exhaust at other isolation boxes in use.
- 2. Proper equipment operation. Since two fan systems are virtually drawing air from the same space the flow direction in the isolation box system could be reversed under certain conditions, such as a fan failure. This means the "cold" to "hot" air flow concept would not be fulfilled. The hottest materials within the isolation boxes would be spread into the cell and into the balcony filters which by virture of their size are much more difficult to dispose of than the box filters within the cell.
- 3. Proper air balancing.
  One central fan is used for 2 cells each of which contains 3 isolation boxes. Unless the air flows are adjusted proper static air pressure conditions may not be realized. The rate at which the box filters become dirty may affect the overall performance even after the system has been balanced.
- 4. Assurance the isolation boxes are under a negative pressure. The plastic material gives a good indication where the net overall air pressure is inside or outside the box, since there is some slack in the material.

## Description of New Facilities

## 1. Improvement of In-Cell Ventilation

Figure No. 2 shows the new design for cells 1 thru 4.

This sketch shows one of the portable ventilators which will built to eliminate the deficiencies of the present systems serving the isolation boxes.

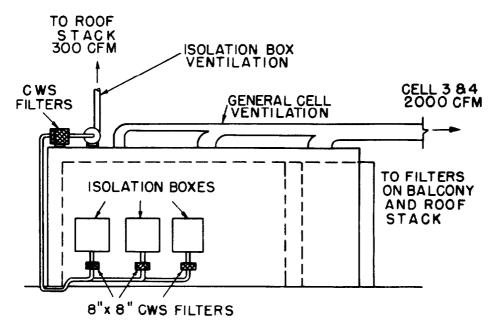


Fig. 1—Cell No. 3, ventilation systems (fan systems also exhaust adjoining Cell No. 4, duplicate of Cell No. 3).

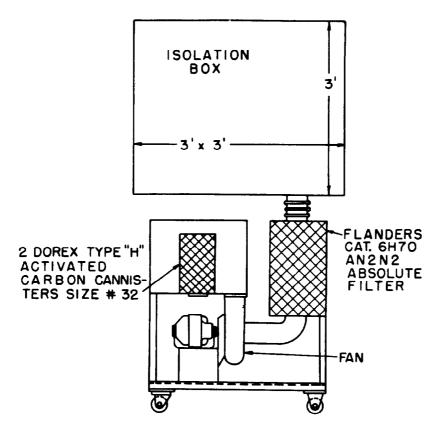


Fig. 2—Portable isolation box vent unit (for Cells 1 through 4).

Each isolation box will be equipped with its own fan and filter as shown. After the air is filtered it will be discharged directly into the cell. The air pressure within the box will always be lower than the cell. Fan failure will not greatly contribute to any spread of materials.

The old isolation box system will be dismantled up to the cell wall and will be for standby use only.

### 2. Central Activated Carbon Filtering System

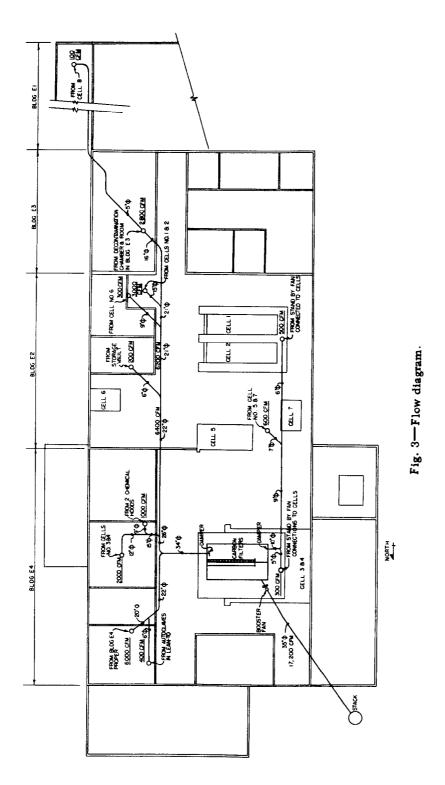
A system of equipment was divised with a view to overcoming the difficiencies outlined.

The first decision related to the redesign of the present systems or to add to them. Except for the in cell isolation box ventilation it was decided to build on to the existing facilities. The reasons were:

- a. We found inadequate in-building space to house additional carbon filters.
- b. We wished to minimize downtime necessary to allow reconstruction of the unit exhaust systems.
- c. It was felt a central system housing all of the carbon filters in a single enclosure will be much more satisfactory from a standpoint of:
  - (1) Shielding, if ever necessary.
  - (2) Dilution of radioactive gases generated at any one of the ventilated work areas as related to the performance of the carbon in filters.
  - (3) A central system will also lend itself to future increased ventilation requirements.
  - (4) Although all the air exhausted will pass thru the carbon filters provision has been made for the installation of a filter by-pass, if we ever desire to install one. Again it is much simpler to install such a by-pass in a central system than it would be with individual unit systems.
- d. Aside from filtering out radioactive iodine discharged from any one of the existing systems, we also desired added protection afforded by a high stack. The stack alone justifies the added ductwork required to interconnect the existing stacks.

The new carbon filtering system is shown in Figure No. 3.

Twelve (12) of the existing ventilation stacks will be cut at the roof line and interconnected with ductwork into two (2) central headers from which air will be drawn by a booster fan thru a filter plenum located on the roof and housing twenty-four (24) "Dorex" Type C, size T-42 cells after which it will be discharged to atmosphere from a 38" diameter, 105 ft. high free standing stack. Except for the stack,



which is to be relocated from the Separations Process Research Unit (no longer in use), all the equipment will be new.

## 3. Safeguard Measures

- a. The individual unit systems equipped with CWS filters ventilate all of the cells and, in addition to this, will provide back-up protection for the portable in-cell CWS-activated carbon units in cells 1 thru 4 where the larger work is done.
- b. Further backup protection will be provided by the central carbon system and stack.
- c. A standby booster fan is to be added. This will start in response to an air flow switch from a separate power source.
- d. Our Health Physics informs us that we can expect an overall radioactive iodine concentration reduction factor in the order of 4 x 10<sup>4</sup> as determined from the following reductions:

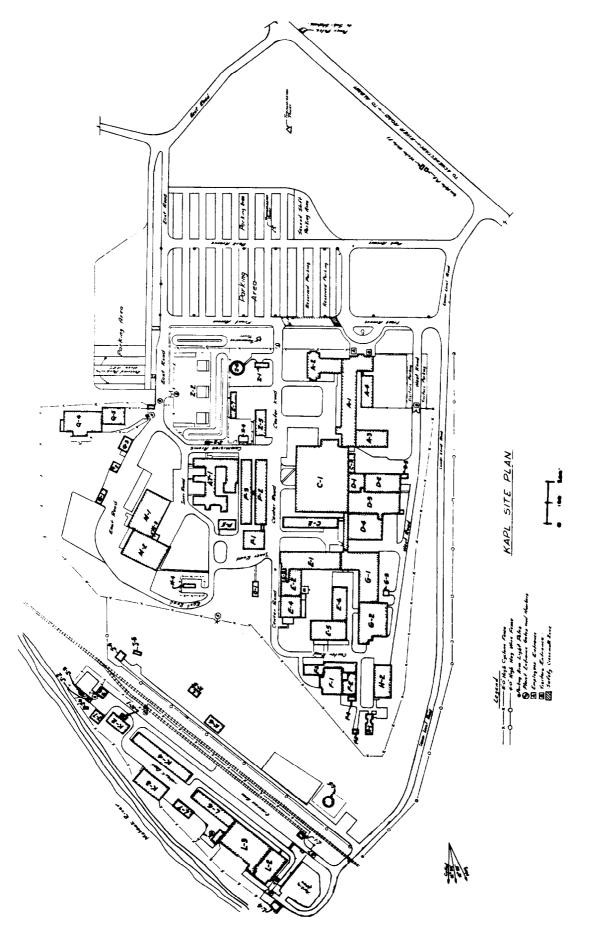
Concentration Reduction	Factor
Stack	100
Activated carbon filters (95% efficient filters will retain up to 18 lbs. radio-active iodine per 24" x 24" filter cell. Two filters in series)	400

e. Provision has been made for the possible addition of a second bank of activated carbon filters in the filter plenum.

overall factor:  $100 \times 400 = 4 \times 10^{4}$ 

- f. The carbon plenum is structurally adequate for future lead shielding up to a thickness of 1/2" along the four walls. The floor of the plenum is masked off by a cell block located immediately below.
- g. Since the adsorption of radioactive xenon and krypton is greatly affected by temperature, the activated carbon will not be relied on for the removal of these gases. The 105foot stack will provide adequate protection here, especially since xenon and krypton do not add to the environs as iodine does.
- h. Highly oxidizing ruthenium tetroxide has been considered. The small amounts which might be given off will separate out in the particulate filters.

In conclusion, the system described should be very satisfactory for the containment of radioactive iodine effluents. Once



Building		Building	
Design		Designation	
A-1	Administration, Engineer. & Technical Offices	K-1	Oil Pump
A-2	Caleteria Physics & Metallurgy Laboratories	7 - Y K- X	ruei uii iank raim Oil Storage Tank
A-4		<b>X</b> -7	Boiler House & Laundry
C-1	General Shops and Offices	<b>х</b> 7-7-	Retention Pits Intermediate Radioactive Waste Storage
2-5	Engineering Offices & Dispensary	,	
ر- د-	Finance Offices	L-1	Hellum Receiving Station Coolant Test Building (Phase I)
D-1	Metallurgy Offices	1 1. 1 6-	Test Building (
D-2	Engineering Department	T-4	
۳ <del>-</del> -	Special Materials Shop (Machining Operations)	η. Ω	
D-5	Metallurgy Snops (Special Materials) Liquid Metals Cleaning Area	L-0 L-1	Engineering Development Radioactive Waste Handling
E-1	Chemistry Laboratoires	M-1	Warehouse
E-2	Radioactive Materials Laboratory	M-2	Warehouse
⊞ :	Chemical Engineering Laboratory	Υ- <b>Σ</b>	Stainless Steel Cutting
E-4	Radioactive Materials Laboratory Addition	M-4	Test Specimen Storage
1 E	Oritical Assemblies Critical Assemblies TRM Wing	[-0	Engineering Offices
)	5 2 2 4 2 5 2 5 5 5 5 5 5 5 5 5 5 5 5 5	<del>!</del>	
F-1	(Preliminar	P-1	
F-12	SIR Proof	P-2	Engineering Offices
7) = E [6	Inysics (Advance Test Reactor)	٠ <u>٠</u> و	brighteering offices
F-5	_	P-5	Records Storage
G-1	Chemistry & Chemical Engineering Laboratories	Q-1	Chemical Storage
G-2	Engineering & Technical Laboratories	9-5	Liquid Metal Storage
<b>G-</b> 3	Scrubber Pump House	<b>6-</b> 3	Site Maintenance Warehouse
		0 4	Development Shops
H-1	Cooling Tower Waste Disnosal Radilities (Timild)	ა-ზ	Development Snops
7.11	ractrictes	2-1	Water and Gas Pump House
J-1		2-2	Gas Storage Traller Stations & Canoples
J-2	Sewage Chlorination	Z-3	Laboratory (Physics)
	Waste Incinerator piitan bad	2-4 7-5	Power Sub-Station Fine House
ታ LC 3 P:	Files. Ded Twhoff Tank	74.	File nouse Water Tower & Valve House
<b>5</b> -15	Injoir Jana Parshall Flume	2-2	3,13,
J-7	Utilities Storage	- !	

the activated carbon system is placed in operation and its operation is observed for a period of time, other data will be made available.

## Discussion

- Q. Have you run any tests?
- A. At present we have one installed as a trial filter been in a matter of two months. going to take measurements yet on this there is only a quarter of an inch of carbon in the cell.
- Q. (J. J. Sabo, U. S. P. H. S. ) I am wondering why your filters are outside of your isolation box, the general tendency has been to put the filter inside the isolation box.
- A. Actually located within seal anyway.
- Q. (J. J. Sabo, U. S. P. H. S. ) When you remove the filter from the isolation box, is there any danger of getting contamination in the cell?
- A. Spread control pretty closely in a cell like this.
- Q. (F. W. Schlapp, Atomics International) The scrubber he described, in it he mentioned that they use 5% solution of sodium hydroxide maintained in this concentration why do they use this solution and how is it maintained, is it by adding water to the solution or some other?
- A. Concentration didn't seem to make any great difference from 2-10% as far as keeping concentrations equal all the time, we don"t, but we do try to maintain it closely.
- Q. (M. D. Thaxter, Lawrence Rad. Lab.) You mentioned your major problem was the dissolver fission in the off-gas handling. I was wondering how much volume was attributable to the process where requirements for dissolving led to the question of whether it would be possible to make a completely closed system dissolution to cut out the major problems?
- A. We have had a maximum of 700 cfm flow about .6 of this would be the normal flow through - new gas products of one sort or another is produced in the dissolving process - newly formed gases or parasitic air -

## ADDITIONAL OFF-GAS FACILITY

A. B. FULLER
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The information contained in this article relays some considerations in the engineering design of additional radioactive off-gas cleaning and handling facilities at Oak Ridge National Laboratory. The new construction program and the goal for ever better decontamination of radioactive effluent has made the need for expanded facilities necessary in the immediate future.

For the past eight months the Engineering Department at Oak Ridge National Laboratory has been actively engaged in the design and the preparation of drawings and specifications for additional radioactive off-gas cleaning facilities. It is expected that these documents will be completed and out for contract bidding within a few weeks. This work will cost in the order of \$230,000 (design costs approximately \$20,000). These added facilities should triple the present gas handling capacity, from 2000 to 6000 cfm.

For the purpose of identification, off gas consists of the waste gases resulting from radioactive processes and from the ventilation of process vessels. Cell ventilation is essentially space ventilation of the over-all space enclosure where the processes are installed.

Present off-gas equipment provides a gas handling capacity of 2000 cfm with cleaning provided by a Cottrell electric precipitator followed by absolute filters with the effluent being released from a 250-foot brick stack along with other gaseous wastes. This system has operated satisfactorily for sometime with the exception of the past six months. During this latter period the gaseous wastes from two process buildings when mixed in the system caused dense smoke (ammonium nitrate) which partially passed the precipitator and quickly caused failure of the absolute filters. This problem has been minimized, but a full solution has not yet been found.

The present normal loading averages approximately 1200 cfm. This normal demand is expected to reach 3 times this level within less than 3 years.

A study of additional off-gas cleaning needs at Oak Ridge National Laboratory dates back over five years with subsequent studies up dating the needs to conform with the changes in the construction program. Oak Ridge National Laboratory is now involved in a construction program having costs in excess of \$20 million, with FYs 60 and 61 planning that adds considerably more. The majority of these projects place an added demand upon the off-gas cleaning facilities.

A decision was made to centralize, as much as possible, all the off-gas cleaning facilities in the central X-10 area to better meet the needs for the service. This means several existing buildings will be served by the equipment planned, or now existing, at the 3039 stack area. The principal existing building to be added is the 3019 Pilot Plant, a chemical processing plant which is allotted as much as 700 cfm flow.

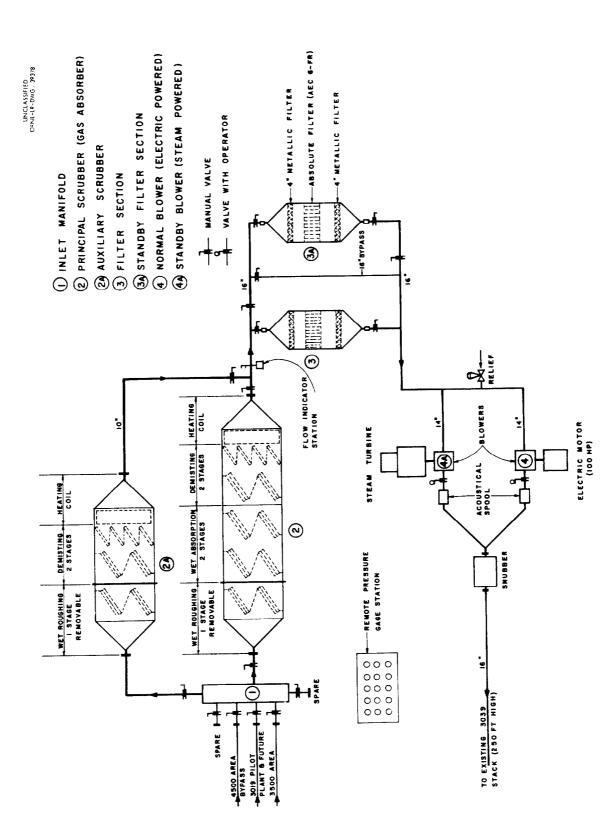
When design work of our new facilities was begun it was immediately evident we needed to know the analysis of the material we were expecting to clean up. A search was initiated to accummulate all the existing available data. For the purposes of design this information turned out to be wholly insufficient. It was not specific, and very little was written fact. To overcome this set-back, we initiated a survey; whereupon all contributors were asked "what gaseous matter do you release to the radioactive off-gas system, how much, at what frequency, what do you expect to do next year, even five years hence." We also sought the advice of the Harvard School of Public Health. In late January of this year, we discussed our problems with Dr. Silverman, Messrs. Dennis, Fitzgerald and others for two days. This visit was very fruitful. However, our survey at Oak Ridge National Laboratory was not successful. Our contributors could not give an accurate and complete analysis of their effluent. The labor, time and other complications in acquiring this information was beyond reason. The nature of operations at Oak Ridge National Laboratory do not include any continuous production where off gas is released. All operations are intermittent, not uniform and in most cases do not exist for long periods of time.

The immediate future will add several new customers to the system among which is the Power Reactor Fuel Reprocessing Facility that is now being designed. This then gives us a list of principal users that includes:

<u>Facility</u>	Max. Flow Allotted
Person Beseton Bush Bernandarian (0507)	(00 0
Power Reactor Fuel Reprocessing (2527)	600 cfm
Multicurie Fission Products Pilot Plant (3517)	600 cfm
3019 Pilot Plant under Chemical Technology Div.	700 c <b>fm</b>
Metal Recovery under Chemical Technology Div.	600 cfm
OR Research Reactor	1000 or more

All uses, both now and within less than the next 3-year period, total over a 4800 cfm maximum flow with normal use expected to average from 60 to 70% of this maximum figure. Twenty-eight or more different buildings or facilities are to be served by the combined systems (old and new). The use of the service varies from a piped fume hood service to high level dissolver venting.

Our final analysis established that the off gas we expect to experience will be (1) quite corrosive, (2) will be radioactive in varying degrees, and (3) will contain only small quantities of particulate matter during normal periods. All particulate is expected to be normally less than 10 microns in size with the major portion less than 1 micron. Short periods of high fume concentrations will result from dissolver reactions.



Schematic routing of gas flow (maximum flow rate, 4,000 scfm.).

This is the worst period of operation believed to be encountered. From this it was decided neutralization must be accomplished before any filtration and, too, the features inherent with wet absorption would best give the required cleaning of both soluble gases and particulate. The selection of stages of absorption were based on trying to achieve 99.5% or better collection of all radioactive iodine for concentrations as low as 5 micrograms per cubic foot of gas. This form of cleaner is believed to be the type best suited to the combined problem of corrosiveness and radioactivity. With a remote underground sump it allows easier shielding of the absorbed activity.

All the principal items of equipment, except blowers, are enclosed with shielding walls with means of performing routine operation without entering the enclosures.

To provide continuous service a steam powered blower and solution pump are included with actuating controls making the changeover automatic. The system will be under the attention of an operator who will make a visual inspection of running equipment and pressure gages each hour. The operator's presence is not required to assist upon the occurrence of a changeover; however, an alarm will sound to indicate this, whereupon the operator can seek out the reason for the change and be aware of the condition.

After considerable investigation and consideration of costs versus life and operation it was decided to fabricate all the ductwork, scrubbers and filter housings of stainless steel. The blowers would be of conventional materials, probably cast iron. Rotary positive blowers were selected to give the more uniform flow over a wide range of pressure drop.

The design requirements for the extensive routing of underground ducts requires a relatively high suction. Up to 50" wg negative pressure is arranged for the inlet box manifold and an additional 30" wg maximum drop is possible through all the cleaning and filtering equipment. Equipment shall be tested airtight and structurally suitable for negative pressure up to 5 psi.

With reference to Drawing 39378 attached, the following will explain the routing of the gas flow.

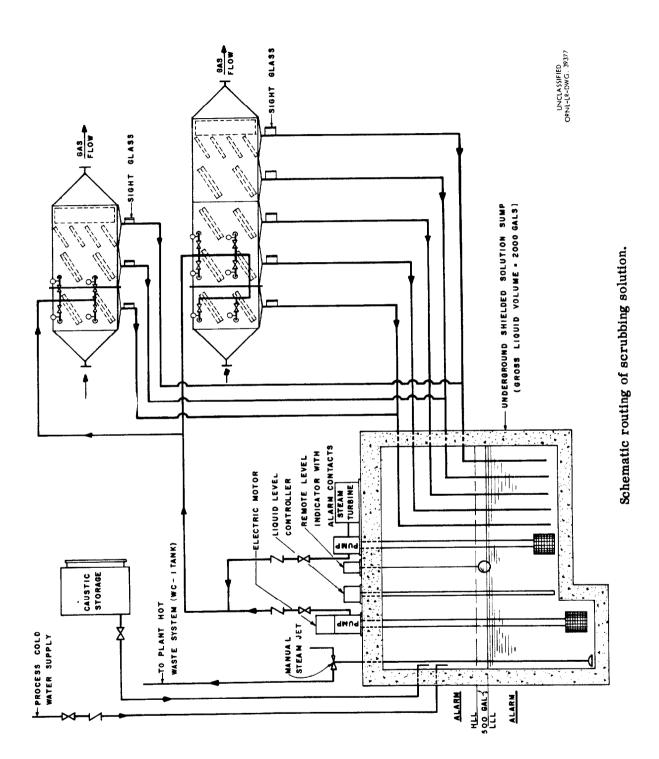
First, we enter the inlet manifold (item #1) on the upper left from the underground collecting duct; then enter the principal scrubber (item #2) which contains the 3 stages of absorption, 2 stages of demisting and finally a steam heating coil to eliminate condensation downstream.

Next, we advance to the dry filter section (item #3), a separate unit from the scrubber. This section contains the 4" metallic filter of silver coated copper, fire resistant absolute filters, and another section of 4" metallic filter, the same material as above.

Following this we pass through the blowers (item #), through the acoustical spool, snubber and on to the existing 3039 stack. This stack is 250 feet high and is used for the release of cell ventilation effluent also. Normally over 120,000 cfm is discharged continuously.

The function of each stage is indicated by pressure differentials on the remote pressure gage station.

The auxiliary scrubber includes one wet roughing stage of absorption followed by two demisting stages and a heating coil. These com-



ponents are equal to the corresponding items in the principal scrubber and are interchangeable. The cell unit assembly is removable as a unit from the top allowing speedy replacement with the minimum of exposure. Spare cell assemblies are planned. The purpose of the two filter sections is to provide continuous service and allow radioactive decay of one section before it is refitted with clean filters. The two are shielded separately.

Effort was made to make possible the easy rearranging of the equipment components. Rather extensive tests are planned for this facility and this flexibility should allow a relative economical means of providing a more efficient system.

Initially the media in the various stages of the scrubber will be:

Wet Roughing: 175-200 micron dia curled Saran or Dynel

Fiber @ 6 to 7 #/cu.ft.

First Wet Absorption: 76-85 micron dia. curled Saran or Dynel Fiber

@ 3.6 to 4 #/cu.ft.

Second Wet Absorption: Same as first wet stage.

First Demisting: Same material as the wet stages but @ 2.8 to

4 #/cu.ft.

Second Demisting: 18-30 micron dia. curled Saran or Dynel Fiber

@ 2.5 to 3 #/cu.ft.

After some use we hope to determine if any plugging or holes occur and correct this by changing the media.

With reference to Drawing 39377 attached, the following will explain the flow of liquid from the underground shielded sump to the scrubber stages and return. The solution is to be 5% sodium hydroxide.

The sump has a gross liquid volume of 2000 gallons; a working volume variation of 500 gallons (or 25%).

Return lines were routed individually to minimize the solution above grade and to afford closer checks of the liquid flow. 3-gpm flow per sq. ft. of media face is set, making a flow to each stage of 66 gpm with a total flow of 198 gpm.

The solution will be periodically sampled, tested and when spent or too radioactive will be jetted manually to the plant hot waste system. Makeup to the sump will be made from the concentrated caustic storage and plant process water system.

The loss of pump discharge pressure initiates the emergency steam powered pump unit.

As mentioned previously extensive tests of this equipment are planned as soon as it is in operation. Perhaps at the next Air Cleaning Seminar a full report of the construction phase, and these tests, can be presented to give conclusive evidence.